The microscopic magnetization: concept and application*

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In this tutorial review a definition of the magnetization which can be used at the microscopic and quantum-mechanical level is motivated and developed. Its implications for the problem of reconstructing the magnetization in a crystal from magnetic neutron-diffraction data or from band-theoretical calculations are discussed, paying special attention to the relation between spin and orbital contributions. [S0034-6861(97)00302-4]

CONTENTS

I. INTRODUCTION

Since the magnetization seemingly should be a fundamental quantity in the description of condensed matter, it is surprising that no consensus has been reached as to how it is to be defined. The beliefs that it is an intrinsically macroscopic field and that it can be determined on the atomic scale by magnetic neutron diffraction somehow manage to coexist in the physics community.

A definition of the magnetization that allows it to be used at the microscopic and quantum-mechanical level in any kind of condensed matter, and without restriction to the quasistatic regime, is in fact available and deserves to be recognized as the fundamental one. It has sometimes implicitly been used in the theory of magnetic neutron scattering and elsewhere, but without ad-

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*Dedicated to Professor Dr. Werner Martienssen.

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equate discussion of its motivation and consequences. To fill that gap is the aim of this tutorial review.

It is well to be clear at the outset as to what new results can and cannot be expected from such a discussion. In the final analysis, the magnetization is only a device for encoding information about the current density, and clarification of this encoding can have no impact on quantities that could be discussed in terms of the current density itself. The prescription for recovering the current density from the magnetization is also unchanged. What does emerge, aside from much-needed conceptual clarification, is new insight into the inverse problem of recovering the magnetization from the current density or from equivalent data such as amplitudes for magnetic neutron scattering.

A formulation that does not suffer from the above shortcomings can be obtained by proceeding in the reverse direction. Instead of trying to deduce in-medium Maxwell equations from suitable definitions of **P** and **M**, one postulates the desired Maxwell equations and asks what definitions of **P** and **M** they imply. The **P** and **M** implied by the Maxwell equations alone are found to be indeterminate by a gauge freedom which is still too large to make them physically useful quantities, but when some of the gauge freedom is eliminated by imposing a suitable further condition, one obtains **P** and **M** with the properties of natural electric and magnetic polarization fields. This formulation is sketched below; a comparison to treatments in the literature and supplementary details are given in Appendices A and B, respectively.

II. ELECTROMAGNETISM IN MEDIA

A. Motivation

The essence of an in-medium formulation of electromagnetism is the use of electric and magnetic polarization fields, **P** and **M**, to generate charge and current densities carried by the medium. These implicit densities can be recovered from **P** and **M** by the well-known relations displayed as Eqs. (2.13) and (2.14) below. Far less easy is the inverse problem of finding suitable **P** and **M** to represent given implicit densities. This is in fact the central difficulty in setting up a satisfactory in-medium formulation.

In the scheme introduced by H. A. Lorentz (1916), **P** and **M** are identified as spatial averages of electric and magnetic dipole moments of the atoms of an insulating medium. For a conducting medium, the densities are first separated into a ‘‘bound’’ part, localized on atoms, and a ‘‘free’’ part, due to conduction electrons or other free charge carriers; the ‘‘bound’’ densities are then represented by **P** and **M** as in an insulator, and the ‘‘free’’ densities are retained as explicit terms in the in-medium Maxwell equations.

Although essentially Lorentzian formulations are still offered in the best-known modern treatises on electromagnetic theory, they are unsatisfactory for two fundamental reasons. First, they make **P** and **M** intrinsically macroscopic by defining them by spatial averaging, but **P** and **M** which can be used at the microscopic level are needed to discuss magnetic neutron scattering and many other topics in the modern theory of condensed matter. Second, because they include no contribution from the orbital current of conduction electrons or other free charge carriers, Lorentzian definitions of **M** are unphysical and incompatible with the definitions used in practice. If the latter difficulty is removed by allowing an orbital contribution to **M** from the free charge carriers, spatial averaging is of no help in finding its **r** dependence. Separation of the densities into ‘‘bound’’ and ‘‘free’’ parts and spatial averaging of the former are then also pointless, so nothing is left of the Lorentzian scheme.

B. The Maxwell equations

The in-vacuum Maxwell equations are

$$\nabla \cdot \mathbf{B} = 0, \tag{2.1}$$

$$c\nabla \times \mathbf{E} + (\partial/\partial t)\mathbf{B} = 0, \tag{2.2}$$

$$\nabla \cdot \mathbf{E} = 4\pi\rho_{\mathrm{tot}}, \tag{2.3}$$

$$c\nabla \times \mathbf{B} - (\partial/\partial t)\mathbf{E} = 4\pi\mathbf{J}_{\mathrm{tot}}. \tag{2.4}$$

In an in-medium formulation, certain charge and current carried by the medium is made implicit. There may also be ‘‘foreign’’ charge and current which is physically distinct from that of the medium, e.g., the current of neutrons scattered by a medium, or a prescribed charge-current distribution whose screening by the medium is to be examined. The most fundamental version of the theory, to be considered first, is that in which only such foreign densities remain explicit. The total charge or current density is then a sum of implicit and foreign parts,

$$\rho_{\mathrm{tot}} = \rho_{\mathrm{impl}} + \rho_{\mathrm{for}}, \tag{2.5}$$

$$\mathbf{J}_{\mathrm{tot}} = \mathbf{J}_{\mathrm{impl}} + \mathbf{J}_{\mathrm{for}}, \tag{2.6}$$

and the in-medium Maxwell equations are[1]

$$\nabla \cdot \mathbf{D} = 4\pi\rho_{\mathrm{for}}, \tag{2.7}$$

$$c\nabla \times \mathbf{H} - (\partial/\partial t)\mathbf{D} = 4\pi\mathbf{J}_{\mathrm{for}}. \tag{2.8}$$

The Maxwell equations imply the continuity equations

$$\nabla \cdot \mathbf{J}_{\mathrm{impl}} + (\partial/\partial t)\rho_{\mathrm{impl}} = 0, \tag{2.9}$$

$$\nabla \cdot \mathbf{J}_{\mathrm{for}} + (\partial/\partial t)\rho_{\mathrm{for}} = 0. \tag{2.10}$$

The Maxwell equations and the identities

$$\mathbf{D} \equiv \mathbf{E} + 4\pi\mathbf{P}, \tag{2.11}$$

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[1] Two further in-medium equations included in Lorentzian formulations, which differ from Eqs. (2.1) and (2.2) only by the insertion of spatial averaging, have no counterparts in the present system.

$$\mathbf{H}\equiv\mathbf{B}-4\pi\mathbf{M}, \qquad (2.12)$$

yield the following expressions for the implicit densities:

$$\rho_{\mathrm{impl}}=-\nabla\cdot\mathbf{P}, \qquad (2.13)$$

$$J_{\mathrm{impl}}=(\partial/\partial t)\mathbf{P}+c\nabla\times\mathbf{M}. \qquad (2.14)$$

No spin terms have been included in the above equations because the internal currents responsible for spin magnetic moments are understood to be included in $\mathbf{J}_{\mathrm{impl}}$. The spin contribution can of course be displayed separately, either as a spin density or as the corresponding spin-carried current density, when necessary.

The above equations apply to any kind of medium. Although they can be understood macroscopically when appropriate, all equations hold microscopically in the first instance. Since the in-vacuum Maxwell equations remain valid when the electromagnetic field is quantized, all equations also hold quantum-mechanically; then **P** and **M**, like the densities ρ_{impl} and $\mathbf{J}_{\mathrm{impl}}$ which they replace, are of course operators.

Postulating the in-medium Maxwell equations is equivalent to imposing Eqs. (2.13) and (2.14) as part of the definitions of **P** and **M**. According to these equations, **P** and **M** imply ρ_{impl} and $\mathbf{J}_{\mathrm{impl}}$ uniquely, but the converse is not true. The nonuniqueness of the polarization fields **P** and **M** for given ρ_{impl} and $\mathbf{J}_{\mathrm{impl}}$ can be expressed as a freedom to redefine them jointly by the gauge transformation

$$\mathbf{P}\rightarrow\mathbf{P}'=\mathbf{P}+\nabla\times\mathbf{G}, \qquad (2.15)$$

$$\mathbf{M}\rightarrow\mathbf{M}'=\mathbf{M}-(1/c)(\partial/\partial t)\mathbf{G}+\nabla g, \qquad (2.16)$$

where $\mathbf{G}(\mathbf{r},t)$ and $g(\mathbf{r},t)$ are arbitrary vector and scalar fields. The fundamental fields **E** and **B** are not affected by this transformation, while the auxiliary fields **D** and **H** are understood to change as implied by $\mathbf{D}\equiv\mathbf{E}+4\pi\mathbf{P}$ and $\mathbf{H}\equiv\mathbf{B}-4\pi\mathbf{M}$.

To arrive at a useful description, one must remove some of this gauge freedom. The first step is to choose between a description by **P** alone and a description by **P** and **M**.

C. Description by P alone

To obtain this description one simply defines

$$\mathbf{M}\equiv 0. \qquad (2.17)$$

Equation (2.14) then reduces to

$$\mathbf{J}_{\mathrm{impl}}\equiv(\partial/\partial t)\mathbf{P}. \qquad (2.18)$$

Formally, there is a residual gauge freedom corresponding to

$$\mathbf{P}\rightarrow\mathbf{P}'=\mathbf{P}+c\nabla\times\mathbf{G}, \qquad (2.19)$$

where $\mathbf{G}(\mathbf{r})$ is an arbitrary time-independent vector field; but since this description is normally used only at finite frequencies, the residual gauge freedom is effectively nil.

The principal advantage of this description is its mathematical economy, which allows the constitutive properties of a linear medium to be described by a dielectric tensor alone, as is familiar from the dielectric theory of metals (Nozières and Pines, 1958; Pines, 1961). Setting $\mathbf{M}\equiv 0$ implies no real loss of information about what would normally be called the magnetic properties, since the magnetic susceptibility tensor of a conventional description can straightforwardly be recovered from the dielectric tensor. Nevertheless, the description by **P** alone is obviously inappropriate when the magnetic properties are of central interest, and so will not be further discussed.

D. Description by P and M

The remainder of this article will employ the description by **P** and **M**, which is preferable for most purposes, especially in view of its closer correspondence to elementary electromagnetic concepts.

1. Definition of P and M

Let subscripts L and T denote the longitudinal and transverse parts of a vector field, which by definition have vanishing curl and divergence, respectively. The transverse part of $\mathbf{J}_{\mathrm{impl}}$ is

$$(\mathbf{J}_{\mathrm{impl}})_T=(\partial/\partial t)\mathbf{P}_T+c\nabla\times\mathbf{M}. \qquad (2.20)$$

The constraint that completes natural definitions of **P** and **M** can be expressed in the equivalent forms

$$(\mathbf{J}_{\mathrm{impl}})_L\equiv(\partial/\partial t)\mathbf{P} \qquad (2.21)$$

and

$$(\mathbf{J}_{\mathrm{impl}})_T\equiv c\nabla\times\mathbf{M}. \qquad (2.22)$$

To motivate this definition, let us first consider the quasistatic case, in which ρ_{impl} is time independent. Then $\mathbf{J}_{\mathrm{impl}}$ is purely transverse and Eq. (2.22) reduces to

$$\mathbf{J}_{\mathrm{impl}}\equiv c\nabla\times\mathbf{M} \quad \text{(quasistatic)}. \qquad (2.23)$$

Partial integration yields

$$\int d^3r\mathbf{M}(\mathbf{r})=(1/2c)\int d^3r\mathbf{r}\times\mathbf{J}_{\mathrm{impl}}(\mathbf{r}). \qquad (2.24)$$

Thus **M** integrates to the total magnetic dipole moment as it should for a natural definition.

The definition of **M** for the dynamic regime is motivated by the requirements that the coupling of the medium to a t-dependent external magnetic field, the Kubo formula for the magnetic susceptibility, and the van Hove formulas for the magnetic neutron-scattering cross sections all be given by expressions in **M** without contributions from $\partial\mathbf{P}/\partial t$, as is natural for nominally magnetic quantities. From results given in Sec. III, it will be clear that each of these criteria is equivalent to a requirement that all the transverse implicit current density be generated by **M**, which is just Eq. (2.22).

2. Gauge freedom of P and M

When Eq. (2.21) or Eq. (2.22) is imposed, the most general gauge transformation that leaves ρ_{impl} and $\mathbf{J}_{\mathrm{impl}}$ invariant is

$$\mathbf{P}\rightarrow\mathbf{P}'=\mathbf{P}+c\nabla\times\mathbf{G}, \tag{2.25}$$

$$\mathbf{M}\rightarrow\mathbf{M}'=\mathbf{M}+\nabla g. \tag{2.26}$$

The joint gauge freedom given by Eqs. (2.15) and (2.16) thus reduces to separate gauge freedoms of **P** and **M**, and the part of ρ_{impl} and $\mathbf{J}_{\mathrm{impl}}$ generated by each field is invariant. Since **P** or **M** can be observed only via the densities it generates, no measurement can determine any particular gauge as the "correct" one. A particular gauge choice may, nevertheless, be preferable if it yields an especially convenient or natural description.

At the macroscopic level it is found empirically that **P** and **M** can usually be described by constitutive equations which give them as functions of the local applied fields. Although gauge-equivalent **P** and **M** satisfying nonlocal constitutive equations usually also exist, a gauge that makes the constitutive equations local is obviously more natural. The requirement that the constitutive equations for **P** and **M** be local fixes, a unique natural gauge for the macroscopic description in most cases, although there is an interesting exception for superconductors (Appendix B.2).

At the microscopic level appropriate choices of gauge within the description by **P** and **M** are very helpful in some contexts, as will be seen in Sec. IV. In other contexts, however, it may be better simply to leave the gauge unspecified.

3. Two-dimensional media

In two-dimensional media, whose practical importance is illustrated by the quantum Hall effect, there is a further simplification: **M** can be taken perpendicular to the medium, and Eq. (2.22) can then be integrated to yield

$$\mathbf{M}(\mathbf{r},t)=(1/c)\int_{\mathbf{r}}^{\infty}d\mathbf{r}'\times\mathbf{J}_{\mathrm{impl}}(\mathbf{r}',t)_{T}, \tag{2.27}$$

where the line integral is along an arbitrary path from the point **r** in the medium to infinity (where **M** vanishes). The gauge freedom is thus eliminated and the uniquely defined **M**(**r**) measures the number of lines of current that must be cut to reach infinity.

E. Medium-carried explicit densities

A modified version of the theory in which parts of the densities carried by the medium are kept explicit is sometimes appropriate. Such medium-carried explicit densities[2] (MCED'S) are needed for a natural description in cases where a static medium carries steady current not due to **M**, since its description by **P** would require **P** to grow linearly in t. This includes all cases where there is a steady net flow through any cross section of the medium, which by Stokes's theorem cannot result from $c\nabla\times\mathbf{M}$.

The medium-carried explicit densities are macroscopic densities defined by constitutive equations such as conduction laws. When they are included, the total densities decompose as

$$\rho_{\mathrm{tot}}=\rho_{\mathrm{impl}}+\rho_{\mathrm{MCED}}+\rho_{\mathrm{for}}, \tag{2.28}$$

$$\mathbf{J}_{\mathrm{tot}}=\mathbf{J}_{\mathrm{impl}}+\mathbf{J}_{\mathrm{MCED}}+\mathbf{J}_{\mathrm{for}}, \tag{2.29}$$

and the in-medium Maxwell equations become

$$\nabla\cdot\mathbf{D}=4\pi(\rho_{\mathrm{MCED}}+\rho_{\mathrm{for}}), \tag{2.30}$$

$$c\nabla\times\mathbf{H}-(\partial/\partial t)\mathbf{D}=4\pi(\mathbf{J}_{\mathrm{MCED}}+\mathbf{J}_{\mathrm{for}}). \tag{2.31}$$

The implicit densities still satisfy the continuity equation (2.9), but the continuity equation (2.10) for the foreign densities is replaced by one for the sum of medium-carried explicit densities and foreign densities,

$$\nabla\cdot(\mathbf{J}_{\mathrm{MCED}}+\mathbf{J}_{\mathrm{for}})+(\partial/\partial t)(\rho_{\mathrm{MCED}}+\rho_{\mathrm{for}})=0. \tag{2.32}$$

Thus medium-carried explicit charge and foreign charge need not be conserved separately. Interconversion between the two necessarily occurs if current flows across the medium's boundary (since medium-carried explicit-densities are naturally required to vanish outside the medium), and conservation of their sum leads to a boundary condition relating the normal components of the two kinds of current density.

After this modified decomposition of the densities, the replacement of the implicit densities by **P** and **M**, which is the essence of the in-medium description, goes through as before. In principle **P** and **M** could still be used microscopically, but only their macroscopic parts are typically of interest in the kind of applications where medium-carried explicit-densities are needed. Medium-carried explicit densities will not arise in the applications discussed in the remainder of this article.

III. MEASUREMENTS INVOLVING THE MICROSCOPIC MAGNETIZATION

The dynamic magnetic susceptibility and the cross section for inelastic magnetic neutron scattering can be expressed in terms of autocorrelations of the magnetization by the Kubo and van Hove formulas (see Secs. III.B and III.C.2, respectively). One would like to prove these important formulas in the greatest possible generality, but their domain of validity can be no larger than that of the definition of **M** used in their proofs. The definition of **M** given in Sec. II.D allows them to be proved in complete generality, i.e., for any kind of condensed matter and with no restriction to the quasistatic regime. The following discussion is intended to explain and justify this assertion without repeating better-known parts of the proofs.

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[2] Medium-carried explicit current density is called "conduction current density" by Landau and Lifshitz (1960), who emphasize that it must not be confused with the total current density carried by the conduction electrons. The somewhat more cumbersome name used here may help to prevent such confusion.

A. Perturbation by external electromagnetic fields

The Kubo and van Hove formulas can be derived most simply by first bringing the coupling between condensed matter and perturbing electromagnetic fields to an appropriate form. In the usual description the Hamiltonian contains electromagnetic potentials ϕ and $\mathbf{A}$ which generate any external fields. The part of the Hamiltonian due to a perturbing part of these fields is

$$H_{\text{per}}=\int d^3r[\rho\phi_{\text{per}}-(1/c)\mathbf{J}\cdot\mathbf{A}_{\text{per}}], \tag{3.1}$$

where ρ and $\mathbf{J}$ are the charge and current densities of the condensed matter. These are the "implicit" densities[3] of Sec. II, which can be reexpressed in terms of $\mathbf{P}$ and $\mathbf{M}$ as discussed there. Equation (3.1) must always hold, since H_{per} simply generates the Lorentz force exerted by the perturbing fields on charged particles in the condensed matter. It can be derived more concretely by writing down the Hamiltonian for a system of interacting charged particles and expanding it to linear order in ϕ_{per} and $\mathbf{A}_{\text{per}}$.

The perturbing potentials are related to perturbing electric and magnetic[4] fields by

$$\mathbf{E}_{\text{per}}=-\nabla\phi_{\text{per}}-(1/c)(\partial/\partial t)\mathbf{A}_{\text{per}}, \tag{3.2}$$

$$\mathbf{H}_{\text{per}}\equiv\mathbf{B}_{\text{per}}=\nabla\times\mathbf{A}_{\text{per}}. \tag{3.3}$$

Expressing the densities in terms of $\mathbf{P}$ and $\mathbf{M}$ and performing partial integrations yields

$$H_{\text{per}}=-\int d^3r[\mathbf{P}\cdot\mathbf{E}_{\text{per}}+\mathbf{M}\cdot\mathbf{H}_{\text{per}}+(1/c)(\partial/\partial t)(\mathbf{P}\cdot\mathbf{A}_{\text{per}})]. \tag{3.4}$$

Separating the fields into longitudinal and transversal parts, and using the fact that integrals of the form $\int d^3r\mathbf{F}_L\cdot\mathbf{G}_T$ vanish (as can easily be seen in the $\mathbf{k}$ representation), yields

$$H_{\text{per}}=-\int d^3r\{\mathbf{P}_L\cdot(\mathbf{E}_{\text{per}})_L+\mathbf{M}_T\cdot\mathbf{H}_{\text{per}}+(1/c)(\partial/\partial t)\times[\mathbf{P}_L\cdot(\mathbf{A}_{\text{per}})_L]+(1/c)[(\partial/\partial t)\mathbf{P}_T]\cdot(\mathbf{A}_{\text{per}})_T\}. \tag{3.5}$$

Defining $\mathbf{P}$ and $\mathbf{M}$ as in Sec. II.D, which implies $(\partial/\partial t)\mathbf{P}_T=0$, and choosing the Coulomb gauge for the electromagnetic potentials, which implies $(\mathbf{A}_{\text{per}})_L=0$, brings the coupling to the final form

$$H_{\text{per}}=-\int d^3r[\mathbf{P}(\mathbf{r},t)_L\cdot\mathbf{E}_{\text{per}}(\mathbf{r},t)_L+\mathbf{M}(\mathbf{r},t)_T\cdot\mathbf{H}_{\text{per}}(\mathbf{r},t)]. \tag{3.6}$$

The applications of present interest involve only the perturbation by a magnetic field,

$$H_{\text{per}}=-\int d^3r\mathbf{M}(\mathbf{r},t)_T\cdot\mathbf{H}_{\text{per}}(\mathbf{r},t). \tag{3.7}$$

B. Kubo formula for magnetic susceptibility

Given Eq. (3.7), the Kubo formula, which expresses the magnetic susceptibility in terms of autocorrelations of the magnetization, can be derived by well-known methods (Kubo, 1957; Kubo *et al.*, 1985):

$$\chi(t-t')_{i\mathbf{k};i'\mathbf{k}'}=i\langle[M_T(\mathbf{k},t)_i,M_T^\dagger(\mathbf{k}',t')_{i'}]\rangle\Theta(t-t'). \tag{3.8}$$

Here $M_T(\mathbf{k},t)_i$ is the Fourier amplitude $\mathbf{k}$ of the Cartesian component i of the transverse part of the magnetization operator and $\Theta(t-t')$ is the unit step function. The derivation of Sec. III.A shows that this result is indeed valid for any kind of condensed matter, microscopically, and without restriction to the quasistatic regime.

C. Magnetic neutron scattering

Results from the theory of magnetic neutron scattering will be reexamined here only insofar as they are related to the definition of the magnetization. More complete reviews, which should be consulted for details omitted below, have been given by Marshall and Lovesey (1971), Lovesey (1984), and Balcar and Lovesey (1989).

1. Electromagnetic coupling of the neutron to condensed matter

In the context of thermal neutron scattering, the neutron can accurately be represented as a point dipole. Identifying the magnetic field $\mathbf{H}_{\text{per}}$ in Eq. (3.7) as that generated by a point-dipole neutron and reexpressing the result in the $\mathbf{k}$ representation yields the following electromagnetic coupling of the neutron to the condensed matter:[5]

$$\langle\mathbf{k}_f|H_{\text{per}}|\mathbf{k}_i\rangle=-4\pi\mathbf{M}(\mathbf{k})_T\cdot(\mathbf{m}_{M1})_n. \tag{3.9}$$

In this expression the coupling is an operator with respect to states of the condensed matter and spin states of the neutron, but its matrix element has been taken between initial and final orbital states of the neutron with momenta $\mathbf{k}_{\text{i}}$ and $\mathbf{k}_f$; $\mathbf{k}\equiv\mathbf{k}_i-\mathbf{k}_f$ is the momentum transfer; $\mathbf{M}(\mathbf{k})_T$ is the Fourier transform of $\mathbf{M}(\mathbf{r})_T$; and $(\mathbf{m}_{M1})_n$ is the dipole-moment operator of the neutron.

2. Van Hove formulas for inelastic scattering

Van Hove (1954) has shown how scattering cross sections in the Born approximation can be rewritten in terms of correlation functions. For thermal neutron scat-

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[3] The subscripts "impl" will be dropped from this point on since other densities will not occur.

[4] Notational conventions for external magnetic fields are discussed in Appendix B.4.

[5] For a careful discussion including some smaller contributions omitted here, see Sears (1986).

tering, the Born approximation is practically exact, and the cross section at momentum and energy transfers $\mathbf{k}$ and ω which follows from the electromagnetic coupling of Eq. (3.9) can be expressed in terms of a dynamic magnetic structure factor

$$S(\mathbf{k},\omega)_{ii'} \equiv \int dt e^{i\omega t} S(\mathbf{k},t)_{ii'}, \tag{3.10}$$

which is the Fourier transform of an autocorrelation of $\mathbf{M}_T$:

$$S(\mathbf{k},t)_{ii'} \equiv \langle M_T(\mathbf{k},t)_i (M_T^\dagger(\mathbf{k})_{i'}\rangle. \tag{3.11}$$

The van Hove formulas for inelastic magnetic neutron scattering, which are given explicitly in Appendix C.1, show how the matrix $S(\mathbf{k},\omega)$ can in principle be completely determined by measuring cross sections with suitable combinations of spin polarization of the incoming neutrons and spin-sensitive detection of the scattered ones. As discussed in Appendix C.2, the dynamic magnetic structure factor is related to the dissipative (anti-Hermitian) part of a one-point reduced susceptibility tensor by

$$S(\mathbf{k},\omega) = [2/(1-e^{-\beta\omega})]^{-1} \mathscr{A}\!\mathscr{h}[\chi_{\mathrm{red}}(\mathbf{k},\omega)]. \tag{3.12}$$

It should be emphasized that these structure factors for inelastic magnetic neutron scattering are valid for any kind of condensed matter and depend on no restriction to the quasistatic regime. The latter restriction would have excluded many applications of interest: for example, inelastic scattering may result from a transition between crystal-field levels of a magnetic ion, which changes the charge density of the ion and thus places the process outside the quasistatic regime.

3. Magnetic Bragg scattering

The coupling of Eq. (3.9) also implies magnetic Bragg scattering amplitudes. Interference between magnetic and nonmagnetic scattering leads to a contribution that depends on the spin polarization of the neutrons, from which values of $\mathbf{u}_{\mathrm{pol}} \cdot \langle \mathbf{M}(\mathbf{K})_T \rangle$ in a magnetically ordered crystal can be deduced. Here $\mathbf{K}$ is the momentum transfer, which for Bragg scattering is a reciprocal lattice vector; $\langle\ \rangle$ is an expectation value at thermal equilibrium; and $\mathbf{u}_{\mathrm{pol}}$ is the axis along which neutron spin is polarized. Since $\langle \mathbf{J}(\mathbf{K}) \rangle = ic\mathbf{K} \times \langle \mathbf{M}(\mathbf{K}) \rangle = ic\mathbf{K} \times \langle \mathbf{M}(\mathbf{K})_T \rangle$, the determination of $\langle \mathbf{M}(\mathbf{K})_T \rangle$ for all $\mathbf{K}$ is equivalent to a complete determination of the current density $\langle \mathbf{J}(\mathbf{r}) \rangle$ in the crystal.

4. Equivalence of spin and orbital contributions

Spin and orbital contributions to magnetic neutron scattering are equivalent in the sense that all scattering formulas can be written in terms of the total (spin plus orbital) magnetization. This equivalence can be traced back to the fact that the neutron "sees" the sample through the electromagnetic coupling only as a current density, making no distinction between currents of spin and orbital origin. The definition of the magnetization discussed in Sec. II.D also refers only to current density and applies to spin and orbital parts equally, so the equivalence carries over to formulas for scattering in terms of the magnetization.

This equivalence means that there is no possibility of a purely experimental separation of spin and orbital contributions in magnetic neutron scattering (or indeed in any purely electromagnetic measurement). One can, however, attempt a heuristic separation with the help of theoretical assumptions about the form of the spin contribution, as will be discussed in Sec. IV.

5. The point-dipolar or small-κ approximation

Early theories of magnetic neutron scattering were plagued by a difficulty in handling the orbital contributions. The difficulty was not in the Born approximation itself, which can straightforwardly be written down in terms of the total current density [the result is just that obtained by substituting Eq. (B1.2) into Eq. (3.11)], but rather in its reexpression via a suitably defined magnetization. Within the Lorentzian conceptual scheme, which was uncritically accepted at the time, a microscopic spin magnetization could be defined via the spin density, but there seemed to be no way to define a microscopic orbital magnetization except by approximating the atom as a point dipole. The spin contribution to magnetic neutron scattering could thus be expressed exactly in terms of the spin magnetization (Bloch, 1936), but a corresponding result for the orbital scattering seemingly could be derived only by making a point-dipolar approximation (Schwinger, 1937).[6] (The point-dipolar approximation is also known as the small-κ approximation because it becomes exact when the momentum transfer, often denoted by κ, is small: see Appendix D.3).

This difficulty disappears when the definition of orbital magnetization corresponding to Eq. (2.22),

$$\mathbf{J}_{\mathrm{orb}}(\mathbf{r},t)_T \equiv c\nabla \times \mathbf{M}_{\mathrm{orb}}(\mathbf{r},t), \tag{3.13}$$

is introduced, as was done for the static case by Trammell (1953) and for the dynamic case by Steinsvoll *et al.* (1967) and Lovesey and Rimmer (1969). The resulting basic formulas for magnetic neutron scattering, which are those already sketched in Secs. III.C.2 and III.C.3 above, are now generally accepted as correct. However, the small-κ approximation had already achieved canonical status, and it somehow escaped attention that the original reason for its use no longer existed. Thus it continues frequently to be used when dealing with orbital magnetization, despite the fact that the condition for its validity, $|\mathbf{k}| r_{\mathrm{ion}} \ll 1$, where r_{ion} is an effective radius of the magnetic ion, is far from being well satisfied in good experiments.[7] Actually there seems to be no need for any such approximation in the more fundamental parts

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[6] In addition to obtaining this approximation for the orbital contribution, Schwinger corrected Bloch's result for the spin contribution, as discussed in Appendix B.4.

[7] It would be practically self-contradictory to assert that the microscopic magnetization could adequately be determined by data satisfying this condition.

of the analysis. So far as magnetic Bragg scattering is concerned, this will be illustrated in Sec. IV.C.

IV. RECONSTRUCTION OF THE MICROSCOPIC MAGNETIZATION IN A CRYSTAL

Let us consider how the thermal expectation value $\langle\mathbf{M}(\mathbf{r})\rangle$ of the microscopic magnetization in a perfect crystal can be reconstructed from the relevant data, which may be magnetic Bragg scattering amplitudes from spin-polarized neutron-diffraction experiments or current and spin densities from band theory. Here "reconstruction" is used to denote a calculational prescription which depends only on the general definition of the magnetization, not on model assumptions, and is thus exact and completely general.

To make such a reconstruction, one must resolve indeterminacies associated with the unboundedness of a perfect crystal and the gauge freedom of the magnetization. Since band theory yields the spin density separately, while neutron-scattering experiments yield only a sum of spin and orbital contributions, the reconstructions needed in the two cases are somewhat different.

A. Indeterminacy due to unboundedness of a perfect crystal

A perfect crystal is either infinite or is made finite by means of periodic boundary conditions; in either case it is unbounded in the sense of having no boundary surface. Band-theoretical calculations are usually made for unbounded crystals, and the sample is effectively infinite in magnetic neutron-diffraction experiments (although not of course in magnetometry measurements).

In unbounded crystals the mean magnetization, i.e., the spatial average of $\langle\mathbf{M}(\mathbf{r})\rangle$, is indeterminate. (If the spin density is known separately, this statement and the cellular construction that follows apply only to the orbital contribution.) Formally, the indeterminacy arises because a constant can be added to $\langle\mathbf{M}(\mathbf{r})\rangle$ without changing the current density[8] $\langle\mathbf{J}(\mathbf{r})\rangle=c\nabla\times\langle\mathbf{M}(\mathbf{r})\rangle$, which is the underlying objective observable. More physically, it arises because the macroscopic current density responsible for the mean magnetization of a uniformly magnetized large finite crystal is localized near the surface, and so is lost in the idealization of an unbounded crystal.

In the limit of "atomic" magnetism due to well-separated magnetic ions, however, the mean magnetization can be recovered by a cellular construction. For simplicity, let the crystal contain a single magnetic ion at each lattice point $\mathbf{R}$. The current density is to be decomposed as

$$\langle\mathbf{J}(\mathbf{r})\rangle\equiv\sum_{R}\langle\mathbf{j}(\mathbf{r}+\mathbf{R})\rangle. \tag{4.1}$$

where the cellular current density $\langle\mathbf{j}(\mathbf{r}+\mathbf{R})\rangle$ is centered at $\mathbf{R}$, but not strictly limited to the corresponding unit cell. If $\langle\mathbf{m}(\mathbf{r})\rangle$ is a cellular magnetization satisfying $\langle\mathbf{j}(\mathbf{r})\rangle=c\nabla\times\langle\mathbf{m}(\mathbf{r})\rangle$, the corresponding magnetization for the crystal is

$$\langle\mathbf{M}(\mathbf{r})\rangle\equiv\sum_{R}\langle\mathbf{m}(\mathbf{r}+\mathbf{R})\rangle. \tag{4.2}$$

The expansion of $\langle\mathbf{j}(\mathbf{k})\rangle$ for small $\mathbf{k}$ is (see Appendix D.3)

$$\langle\mathbf{j}(\mathbf{k})\rangle=ic\mathbf{k}\times\langle\mathbf{m}_{M1}\rangle+O(k^2), \tag{4.3}$$

where

$$\langle\mathbf{m}_{M1}\rangle\equiv(1/2c)\int d^3r\mathbf{r}\times\langle\mathbf{j}(\mathbf{r})\rangle$$
$$=\int d^3r\langle\mathbf{m}(\mathbf{r})\rangle=\int_{[\mathrm{cell}]}d^3r\langle\mathbf{M}(\mathbf{r})\rangle \tag{4.4}$$

is the magnetic dipole moment of the cellular current. To recover $\langle\mathbf{m}_{M1}\rangle$ and hence the mean magnetization, one must recover $\langle\mathbf{j}(\mathbf{r})\rangle$ from the given $\langle\mathbf{J}(\mathbf{r})\rangle$. Fourier transformation of Eq. (4.1) yields

$$\langle\mathbf{j}(\mathbf{K})\rangle=\langle\mathbf{J}(\mathbf{K})\rangle \tag{4.5}$$

for any reciprocal-lattice vector $\mathbf{K}$, but $\langle\mathbf{j}(\mathbf{k})\rangle$ for $\mathbf{k}\neq\mathbf{K}$ is formally indeterminate except for the stationary-flow constraint

$$\mathbf{k}\cdot\langle\mathbf{j}(\mathbf{k})\rangle=0. \tag{4.6}$$

The definition of $\langle\mathbf{j}(\mathbf{k})\rangle$ must thus be completed by a smooth interpolation through the given values at $\mathbf{k}=\mathbf{K}$, which is a standard problem in numerical analysis.

For atomic magnetism $\langle\mathbf{J}(\mathbf{r})\rangle$ is well localized near the lattice points. The Fourier amplitudes $\langle\mathbf{J}(\mathbf{K})\rangle$ then remain significant out to $|\mathbf{K}|\gg|\mathbf{K}|_{\min}$ and effectively form a continuum, allowing the cellular current and hence the mean magnetization to be identified unambiguously. For increasingly less well localized $\langle\mathbf{J}(\mathbf{r})\rangle$, any determination of the mean magnetization by data pertaining to the unbounded crystal becomes increasingly uncertain. [One can of course easily arrange for the interpolated $\langle\mathbf{j}(\mathbf{k})\rangle$ to reproduce a value of the mean magnetization that might be known from magnetometry.]

B. Indeterminacy due to gauge freedom of the magnetization

As discussed in Sec. II, the magnetization is subject to an intrinsic gauge freedom, which allows it to be redefined by

$$\langle\mathbf{M}(\mathbf{r})\rangle\rightarrow\langle\mathbf{M}'(\mathbf{r})\rangle=\langle\mathbf{M}(\mathbf{r})\rangle+\nabla g(\mathbf{r}), \tag{4.7}$$

meaning that its longitudinal part $\langle\mathbf{M}(\mathbf{r})_L\rangle$ can be chosen arbitrarily. Despite the work of Brown *et al.* (1973) and Balcar (1975), there has been insufficient awareness of the existence of such a gauge freedom and inadequate understanding of how to deal with it when reconstructing $\langle\mathbf{M}(\mathbf{r})\rangle$ in a crystal.

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[8] We shall be concerned only with the thermal-average current density, which is stationary and hence purely transversal: $\langle\mathbf{J}(\mathbf{r})\rangle\equiv\langle\mathbf{J}(\mathbf{r})_T\rangle$.

In principle, no gauge need be selected for the magnetization, since it could be described by tabulating $\langle\mathbf{M}(\mathbf{K})\rangle$, with values of the component parallel to $\mathbf{K}$, which is the longitudinal part, simply left undefined. However, in order to plot $\langle\mathbf{M}(\mathbf{r})\rangle$, as is often useful when discussing neutron-scattering experiments, some gauge must be chosen. Different gauges may yield quite different plots and an appropriate gauge can help to bring out the physics.

1. The crystalline Trammell gauge

One of the things one might want to learn from a plot of $\langle\mathbf{M}(\mathbf{r})\rangle$ is whether the magnetism is of atomic type. The "solenoidal" gauge defined by setting $\langle\mathbf{M}(\mathbf{r})_L\rangle\equiv 0$, although formally an obvious choice, is unsuitable for this purpose, since it makes $\langle\mathbf{M}(\mathbf{r})\rangle$ proportional to the $\mathbf{B}$ field generated by $\langle\mathbf{J}(\mathbf{r})\rangle$, which is poorly localized even when $\langle\mathbf{J}(\mathbf{r})\rangle$ is well localized [e.g., a magnetic ion with an exponentially bounded $\langle\mathbf{j}(\mathbf{r})\rangle$ would be described by an $\langle\mathbf{m}(\mathbf{r})\rangle$ falling off only as r^{-3} at large $\mathbf{r}$].

A definition yielding a well-localized ionic $\langle\mathbf{m}(\mathbf{r})\rangle$ is that of Trammell (1953):

$$\langle\mathbf{m}(\mathbf{r})\rangle\equiv(1/c)\int_1^\infty d\lambda\,\lambda\,\mathbf{r}\times\langle\mathbf{j}(\lambda\mathbf{r})\rangle. \tag{4.8}$$

With the origin taken at the center of the ion, this formula places each contribution radially inward from the current that generates it, thus indeed yielding a well-localized $\langle\mathbf{m}(\mathbf{r})\rangle$, which can be shown to satisfy $c\nabla\times\langle\mathbf{m}(\mathbf{r})\rangle=\langle\mathbf{j}(\mathbf{r})\rangle$ (see Appendix D.4) and thus constitutes a legitimate gauge choice.

Because Eq. (4.8) depends on the choice of origin, applying it directly to a crystal as a whole would yield a magnetization without the translation symmetry[9] of the crystal. An $\langle\mathbf{M}(\mathbf{r})\rangle$ with the appropriate symmetry can, however, be constructed by forming a cellular $\langle\mathbf{j}(\mathbf{k})\rangle$ as discussed above, defining a cellular $\langle\mathbf{m}(\mathbf{r})\rangle$ by Eq. (4.8), and superposing the latter by Eq. (4.2).

The "crystalline Trammell gauge" defined by the above procedure is a theoretically interesting choice for the reconstruction of the magnetization, but the alternative choice to be discussed next is more useful in practice because it is computationally simpler and matches up better with the analysis of magnetic Bragg scattering of neutons.

2. The most nearly collinear (MNC) gauge

For spin magnetization, the natural gauge from a theoretical point of view is the "spin-density" gauge defined by

$$\langle\mathbf{M}_{\text{spin}}(\mathbf{r})\rangle\equiv g_0\mu_A\langle\mathbf{s}(\mathbf{r})\rangle, \tag{4.9}$$

where $\langle\mathbf{s}(\mathbf{r})\rangle$ is the spin density and the other quantities are defined in Appendix D.1. Although the spin density is completely observable in principle, only its transverse part can be determined by magnetic neutron diffraction or other purely electromagnetic measurements (see Appendix D.1). Even assuming orbital contributions to be negligible, the experimentalist can thus hope to recover from neutron-scattering data the same $\langle\mathbf{M}_{\text{spin}}(\mathbf{r})\rangle$ that the theorist would regard as natural only with the help of an educated guess as to its gauge.

Such a guess may be based on the fact that the spin density is everywhere collinear in the simplest shell-model states of an isolated atom, as well as in the simplest theories of itinerant ferromagnetism. This suggests defining a "most nearly collinear" (MNC) gauge by

$$\int d^3r\,|\langle\mathbf{M}(\mathbf{r})\rangle_\perp|^2=\text{minimum}, \tag{4.10}$$

where $\langle\mathbf{M}(\mathbf{r})\rangle_\perp$ is the part of $\langle\mathbf{M}(\mathbf{r})\rangle$ perpendicular[10] to a unit vector $\mathbf{u}_\parallel$, which may be chosen as the direction $\mathbf{u}_{\mathbf{M1}}$ of the mean magnetization or as the direction of a sublattice magnetization or external magnetic field. The minimum is to be obtained by varying $\langle\mathbf{M}(\mathbf{r})_L\rangle$, with $\langle\mathbf{M}(\mathbf{r})_T\rangle$ fixed by the current density.

In the $\mathbf{K}$ representation Eq. (4.10) becomes

$$|\langle\mathbf{M}(\mathbf{K})\rangle_\perp|^2=\text{minimum}\,\forall\mathbf{K}. \tag{4.11}$$

Since the freedom of $\langle\mathbf{M}(\mathbf{K})_L\rangle$ means that an arbitrary part parallel to $\mathbf{K}$ can be added to $\langle\mathbf{M}(\mathbf{K})\rangle$, the minimum is achieved when

$$\mathbf{K}_\perp\cdot\langle\mathbf{M}(\mathbf{K})\rangle_\perp=0\,\forall\mathbf{K}. \tag{4.12}$$

From $\langle\mathbf{J}(\mathbf{r})\rangle=c\nabla\times\langle\mathbf{M}(\mathbf{r})\rangle$ follows

$$\langle\mathbf{M}(\mathbf{K})_T\rangle=(i/c)\mathbf{K}\times\langle\mathbf{J}(\mathbf{K})\rangle/\mathbf{K}\cdot\mathbf{K}. \tag{4.13}$$

The solution for the MNC magnetization in terms of the current density implied by the last two equations is (Appendix D.5)

$$\langle\mathbf{M}(\mathbf{K})\rangle=(i/c)\mathbf{K}_\perp\times\langle\mathbf{J}(\mathbf{K})\rangle/\mathbf{K}_\perp\cdot\mathbf{K}_\perp\quad(\mathbf{K}_\perp\neq 0). \tag{4.14}$$

If reexpressed in terms of $\langle\mathbf{M}(\mathbf{K})_T\rangle$, the quantity immediately yielded by magnetic Bragg scattering, the solution reads

$$\langle\mathbf{M}(\mathbf{K})\rangle=(\mathbf{K}\times\langle\mathbf{M}(\mathbf{K})_T\rangle)\times\mathbf{K}_\perp/\mathbf{K}_\perp\cdot\mathbf{K}_\perp\quad(\mathbf{K}_\perp\neq 0). \tag{4.15}$$

In agreement with the earlier discussion, $\langle\mathbf{M}(\mathbf{K})\rangle$ at $\mathbf{K}=\mathbf{0}$ (the mean magnetization) is indeterminate except inso-

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[9] Here the principle that the gauge freedom should be restricted so as to admit only $\langle\mathbf{M}(\mathbf{r})_L\rangle$ of the correct symmetry is being invoked. The symmetry is defined unambiguously by the current density.

[10] The parts $\mathbf{M}_\parallel$ and $\mathbf{M}_\perp$ parallel and perpendicular to the fixed $\mathbf{u}_\parallel$ must not be confused with the longitudinal and transverse parts $\mathbf{M}_L$ and $\mathbf{M}_T$, which are the parts parallel and perpendicular to the variable $\mathbf{K}$ in the $\mathbf{K}$ representation and which are also sometimes denoted $\mathbf{M}_\parallel$ and $\mathbf{M}_\perp$ in the literature.

far as it can be recovered from a cellular construction. For $\mathbf{K}\neq\mathbf{0}$, $\mathbf{K}_\perp=0$, $\langle\mathbf{M}(\mathbf{K})\rangle_\perp$ is fixed by Eq. (4.13), but $\langle\mathbf{M}(\mathbf{K})\rangle_\parallel$ is indeterminate. Natural values for $\langle\mathbf{M}(\mathbf{K})\rangle_\parallel$ at such $\mathbf{K}$ can be assigned by interpolation (Appendix D.5).

This very simple solution, which for generic $\mathbf{K}$ gives $\langle\mathbf{M}(\mathbf{K})\rangle$ in terms of $\langle\mathbf{J}(\mathbf{K})\rangle$ or $\langle\mathbf{M}(\mathbf{K})_T\rangle$ at the same $\mathbf{K}$, may be contrasted with the solution in the crystalline Trammell gauge, which is nonlocal in $\mathbf{K}$ as a result of the indirect construction via a cellular current density.

How can the minimal noncollinear magnetization that remains in the MNC gauge be interpreted? As noted at the outset, it is usually possible to describe spin contributions by a parallel magnetization in good approximation. Since $\langle\mathbf{J}(\mathbf{r})\rangle\equiv c\nabla\times\langle\mathbf{M}(\mathbf{r})\rangle$, a perfectly parallel orbital magnetization can be found only if the orbital current density lies entirely in planes perpendicular to $\mathbf{u}_{M1}$. This is true for an ion in free space in a standard angular-momentum eigenstate; but as such ions are brought together to form a crystal, their orbital current density, which is more sensitive to the environment than is the spin, tends to be warped out of plane, first by crystal-field effects and then by band-structure effects. Thus the noncollinear part of $\langle\mathbf{M}(\mathbf{r})\rangle$ in the MNC gauge can be attributed primarily to orbital contributions and may serve as an "earmark" to detect them.[11]

C. Analysis of magnetic Bragg scattering data

The results developed in the preceding discussion are helpful in the analysis of magnetic Bragg scattering of neutrons, where they allow exact evaluation of certain quantities whose evaluation had previously seemed to depend on approximations. The sketch that follows emphasizes the points where the definition of the magnetization is essential and is not intended as a well-rounded review of such analyses. A brief survey of the relevant literature is given in Appendix E, and further details can be found in the treatises cited at the beginning of Sec. III.C.

1. Experimental limitations

A complete reconstruction of the microscopic magnetization is possible only when the current density, or equivalently the transverse part of the magnetization, is completely known. For magnetic Bragg scattering, this would mean measurement of all three components of $\langle\mathbf{M}(\mathbf{K})_T\rangle$ for all magnetic reciprocal-lattice vectors $\mathbf{K}$, which is not possible in view of the following experimental limitations:

(1) Measurements are feasible only for values of $\mathbf{K}$ that are not too large.

(2) For ferromagnetic crystals an external magnetic field is needed to suppress formation of magnetic domains, and it is difficult if not impossible to polarize the neutrons except along the direction of the field. This means that only $\langle\mathbf{M}(\mathbf{K})_T\rangle_\parallel$, the component of $\langle\mathbf{M}(\mathbf{K})_T\rangle$ parallel to the field, can be determined from the nuclear-magnetic interference term in the Bragg scattering of polarized neutrons. One can also determine $|\langle\mathbf{M}(\mathbf{K})_T\rangle|^2$ by nonpolarized Bragg scattering and then attempt to find $|\langle\mathbf{M}(\mathbf{K})_T\rangle_\perp|^2$ by subtracting $|\langle\mathbf{M}(\mathbf{K})_T\rangle_\parallel|^2$, but this is difficult when $|\langle\mathbf{M}(\mathbf{K})_T\rangle_\perp|$ is small, as will often be the case.

(3) For antiferromagnetic crystals, the amplitudes $\langle\mathbf{M}(\mathbf{K})_T\rangle$ for $\mathbf{K}$ which belong to the chemical reciprocal lattice tend to be small and often vanish by symmetry. The amplitudes for $\mathbf{K}$ that belong to the magnetic but not the chemical reciprocal lattice cannot be determined from the nuclear-magnetic interference term, but the absolute value of any vector component of $\langle\mathbf{M}(\mathbf{K})_T\rangle$ can, in principle, determined from the purely magnetic Bragg scattering of suitably polarized neutrons.

Limitation 1 is not too serious, since one can hope to measure at enough $\mathbf{K}$ to allow a plausible extrapolation to arbitrarily large $\mathbf{K}$, and it will be ignored in what follows. The remaining limitations pose greater difficulties and seemingly make an analysis impossible except with the help of approximations or model assumptions.

2. Analysis for a ferromagnetic crystal

A model assumption that immediately suggests itself for ferromagnetic crystals, and that allows analysis of Bragg scattering data, is that the microscopic magnetization is perfectly collinear[12] and parallel to the external field:

$$\langle\mathbf{M}(\mathbf{r})\rangle=\langle\mathbf{M}(\mathbf{r})\rangle_\parallel;\quad \langle\mathbf{M}(\mathbf{r})\rangle_\perp=0. \tag{4.16}$$

The general expression for the transverse magnetization in the $\mathbf{K}$ representation is

$$\langle\mathbf{M}(\mathbf{K})_T\rangle\equiv\langle\mathbf{M}(\mathbf{K})\rangle-\mathbf{K}\mathbf{K}\cdot\langle\mathbf{M}(\mathbf{K})\rangle/\mathbf{K}\cdot\mathbf{K}. \tag{4.17}$$

When Eq. (4.16) holds, the parallel part of $\langle\mathbf{M}(\mathbf{K})_T\rangle$, whose measurement is unproblematic, is

$$\langle\mathbf{M}(\mathbf{K})_T\rangle_\parallel=\langle\mathbf{M}(\mathbf{K})\rangle-\mathbf{K}_\parallel\cdot\mathbf{K}_\parallel\langle\mathbf{M}(\mathbf{K})\rangle/\mathbf{K}\cdot\mathbf{K} \tag{4.18}$$

and $\langle\mathbf{M}(\mathbf{K})\rangle=\langle\mathbf{M}(\mathbf{K})\rangle_\parallel$ can be determined as

$$\langle\mathbf{M}(\mathbf{K})\rangle_\parallel=(\mathbf{K}\cdot\mathbf{K}/\mathbf{K}_\perp\cdot\mathbf{K}_\perp)\langle\mathbf{M}(\mathbf{K})_T\rangle_\parallel. \tag{4.19}$$

The magnetization[13] $\langle\mathbf{M}(\mathbf{r})\rangle=\langle\mathbf{M}(\mathbf{r})\rangle_\parallel$ can thus be found from the Bragg scattering data, but apparently only in the approximation that it is perfectly collinear.

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[11] Although for the simplest magnetic systems noncollinear magnetization in the MNC gauge tends to arise primarily via orbital contributions, it cannot conversely be concluded that orbital magnetization in such systems is mainly noncollinear. On the contrary, in typical examples, such as those discussed in Appendix D.6, the collinear part of the MNC orbital magnetization is still larger than the noncollinear part.

[12] To the extent that the crystal can be approximately described by magnetic dipole moments of point ions, these are parallel by definition in the ferromagnetic state, but in a continuum description there is no requirement that $\langle\mathbf{M}(\mathbf{r})\rangle$ be collinear.

[13] The expression "magnetization density," which is often used to denote this quantity, should be rejected as a pleonasm.

Very remarkably, this result actually holds exactly if the MNC gauge is adopted. As shown in Appendix D.5, in that gauge the general solution for $\langle \mathbf{M}(\mathbf{K})\rangle$ can be expressed as

$$\langle \mathbf{M}(\mathbf{K})\rangle = (\mathbf{K}\cdot\mathbf{K}/\mathbf{K}_\perp\cdot\mathbf{K}_\perp)\langle \mathbf{M}(\mathbf{K})_T\rangle_\parallel + \langle \mathbf{M}(\mathbf{K})_T\rangle_W \quad (\mathbf{K}_\perp \neq 0). \tag{4.20}$$

Here the $\perp$ subspace has been separated into a part along the direction $\mathbf{v}$ of $\mathbf{K}_\perp$ and a part along $\mathbf{w}\equiv\mathbf{u}_\parallel\times\mathbf{v}$. The part of $\langle \mathbf{M}(\mathbf{K})\rangle$ along $\mathbf{v}$ is identically zero, the part along $\mathbf{w}$ equals the corresponding part of $\langle \mathbf{M}(\mathbf{K})_T\rangle$, and the parallel part agrees with Eq. (4.19). Use of the MNC gauge has arranged things so that $\langle \mathbf{M}(\mathbf{K})_T\rangle_\perp$, which cannot conveniently be observed, affects only $\langle \mathbf{M}(\mathbf{K})\rangle_W$. Thus an exact reconstruction of at least the parallel[14] component of $\langle \mathbf{M}(\mathbf{r})\rangle$ from data of the kind yielded by practical scattering experiments is possible after all.

It should be emphasized that this conclusion holds for atomic, itinerant, or any intermediate kind of magnetism, and with arbitrarily large orbital contributions.

3. Analysis for an antiferromagnetic crystal

For simplicity, the present discussion will be limited to the case of a "bipartite" antiferromagnet, ordered by sublattices A and B which are isomorphic and contain each other's nearest neighbors. By symmetry $\langle \mathbf{M}(\mathbf{K})\rangle$ vanishes for $\mathbf{K}$ that belong to the chemical reciprocal lattice and is real for $\mathbf{K}$ that do not. Thus the indeterminacy of the values of $\langle \mathbf{M}(\mathbf{K})_T\rangle$ that can be obtained from neutron scattering (see limitation 3 above) reduces to an unknown sign. If model assumptions are available from which this sign can confidently be deduced, $\langle \mathbf{M}(\mathbf{r})\rangle$ can be exactly evaluated from data of the kind yielded by practical scattering experiments (although this is not a "reconstruction" in the strict sense defined at the outset). The MNC gauge (with $\parallel$ defined by the common axis of the moments on the two sublattices) is still appropriate, and it now becomes possible to determine $\langle \mathbf{M}(\mathbf{r})\rangle_\perp$ and thus to use it as an "earmark" for orbital contributions.

V. CONCLUSIONS

Although a definition of the magnetization that can be used at the microscopic level is needed for the analysis of magnetic neutron scattering and for many other problems in the modern theory of condensed matter, there has been a general reluctance to examine the necessary definition closely and formulate it precisely. This seems to be due in part to a lingering belief, inherited from Lorentzian theories of electromagnetism in media, that a true magnetization can be defined only macroscopically. That belief is false. A definition is in fact available which applies microscopically and quantum mechanically, with no restriction to the quasistatic regime, and which can be motivated very soundly. It relates the magnetization to the transverse part of the medium-carried implicit current density by Eq. (2.22):

$$(\mathbf{J}_{\text{impl}})_T = c\nabla\times\mathbf{M}.$$

The magnetization yielded by this definition is subject to a gauge freedom which allows its longitudinal part to be redefined arbitrarily, implying that only its gauge-invariant transverse part can be determined objectively. The well-known fact that only the transverse part of the magnetization is observed in magnetic neutron scattering is a particular instance of this more general principle.

Spin and orbital contributions enter the magnetization on an equal footing, meaning that it is impossible to distinguish rigorously the two by magnetic neutron scattering or any other purely electromagnetic measurement. More heuristically, however, it may be possible to detect orbital contributions by invoking a tendency of spin contributions to be describable by a perfectly collinear magnetization.

In order to reconstruct the microscopic magnetization from band theory or neutron-scattering experiments, some definite gauge must be adopted. A "most nearly collinear" gauge is found to be useful for the analysis of magnetic Bragg scattering, where it allows the part of the magnetization parallel to the orienting external field acting on a ferromagnetic crystal to be reconstructed exactly from data of the kind yielded by practical experiments with polarized neutrons. This reconstruction is valid for atomic, itinerant, or any intermediate kind of magnetization and with arbitrarily large orbital contributions. With the help of minimal model assumptions, related exact determinations are also possible for antiferromagnets.

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APPENDIX A: ELECTROMAGNETISM IN MEDIA—LITERATURE

The specialized literature on electromagnetic media is made up primarily of neo-Lorentzian approaches. The original work of Lorentz (1916) employed prescriptions for spatial averaging that were not entirely satisfactory, and subsequent work (de Groot, 1969; Russakoff, 1970; de Groot and Suttorp, 1972; Robinson, 1973) has achieved great improvements in that area. Unfortunately, the more fundamental difficulty mentioned in Sec. II.A, namely the missing contribution to $\mathbf{M}$ from the orbital current of conduction electrons or other free

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[14] If the directions of the neutron polarization and the mean magnetization are different (Wulff *et al.*, 1988), the former must be identified as the $\parallel$ direction for the MNC gauge in order for this result to hold.

charge carriers, which somehow escaped attention, makes such formulations unacceptable.

The unphysical Lorentzian definition of **M** for conducting media never leads to unphysical results because it is never really used. It is invoked to obtain a supposed justification of the in-medium Maxwell equations, but is tacitly replaced by a more realistic definition for practical work. No experimentalist has ever determined the magnetic equation of state of a conductor by measuring the dipole moment of a sample and then subtracting off an estimated orbital contribution from the conduction electrons to obtain a corrected moment corresponding to the "true" magnetization, as the Lorentzian definition would require him to do.

Landau and Lifshitz (1960) recognized the above difficulty and emphasized that **M** must, in general, include an orbital contribution from conduction electrons or other free charge carriers. However, they continued to accept the Lorentzian idea that **P** and **M** are intrinsically limited to the macroscopic (and also quasistatic) regime.

Post (1971) and Kovetz (1990) (see also Jelitto, 1994) recognized that definition of **P** and **M** in terms of the underlying charge and current densities implies that they are subject to the gauge freedom of Eqs. (2.15) and (2.16), but they again assumed these fields to be inherently macroscopic and so failed to arrive at the microscopically valid definition of the magnetization obtained here.

APPENDIX B: ELECTROMAGNETISM IN MEDIA—SUPPLEMENTARY DISCUSSION

1. Relations between magnetization and current density

The magnetization was defined in the **r** representation by

$$\mathbf{J}_{\mathrm{impl}}(\mathbf{r})_T = c\nabla\times\mathbf{M}(\mathbf{r}),$$

which fixes $\mathbf{M}(\mathbf{r})_T$ uniquely while leaving $\mathbf{M}(\mathbf{r})_L$ arbitrary.

It is often convenient to work in the **k** representation, where an explicit expression for $\mathbf{M}(\mathbf{k})_T$ in terms of $\mathbf{J}(\mathbf{k})_T$ is available:

$$\mathbf{J}_{\mathrm{impl}}(\mathbf{k})_T \equiv ic\mathbf{k}\times\mathbf{M}(\mathbf{k}) \equiv ic\mathbf{k}\times\mathbf{M}(\mathbf{k})_T, \tag{B1.1}$$

$$\mathbf{M}(\mathbf{k})_T \equiv [\mathbf{k}\times\mathbf{M}(\mathbf{k})]\times\mathbf{k}/\mathbf{k}\cdot\mathbf{k} \equiv ic\mathbf{k}\times\mathbf{J}_{\mathrm{impl}}(\mathbf{k})_T/(ck)^2. \tag{B1.2}$$

The singularity at **k**=0 in the last expression is related to the fact that in the static case, $\mathbf{M}(\mathbf{r})_T$ is proportional to the magnetostatic **B** field generated by $\mathbf{J}_{\mathrm{impl}}(\mathbf{r})$, which has a long-range dipolar tail unless the dipolar moment of the current distribution $\mathbf{J}_{\mathrm{impl}}(\mathbf{r})$ happens to vanish. This singularity causes no difficulties if treated with appropriate care.

2. The natural gauge for macroscopic magnetization

Empirically, it is found that magnetometry data are usually consistent with a constitutive equation

$$\mathbf{M}_{\mathrm{mac}}(\mathbf{r}) = \mathbf{M}_{\mathrm{mac}}[\mathbf{H}_{\mathrm{mac}}(\mathbf{r})], \tag{B2.1}$$

according to which the macroscopic magnetization is a function of the local macroscopic **H** field. This is obviously the natural description. Nevertheless, one could in principle construct a gauge-equivalent alternative $\mathbf{M}_{\mathrm{mac}}$ by retaining the old curl, while assigning new values to the divergence.[15]

In general such a gauge transformation would scramble the description in such as way as to make the magnetic constitutive equation nonlocal; but an exception occurs for simply connected[16] Type-I superconductors in sufficiently low fields. The constitutive equation is then $\mathbf{B}_{\mathrm{mac}}=0$, which can be rewritten in the form of Eq. (B2.1) as

$$\mathbf{M}_{\mathrm{mac}}(\mathbf{r}) = (-1/4\pi)\mathbf{H}_{\mathrm{mac}}(\mathbf{r}). \tag{B2.2}$$

Since the **B** field is invariant under gauge transformations, the new **M** and **H** fields continue to satisfy the old constitutive equation. Thus the requirement that the constitutive equation be local does not yet fix the gauge of $\mathbf{M}_{\mathrm{mac}}(\mathbf{r})$. An appropriate condition to complete the fix of a natural macroscopic gauge in this exceptional case is

$$\nabla\cdot\mathbf{M}_{\mathrm{mac}}(\mathbf{r}) = 0 \quad \text{(inside sample)}, \tag{B2.3}$$

which for an ellipsoidal sample in a uniform external field yields the familiar simple solution in which $\mathbf{M}_{\mathrm{mac}}$ and $\mathbf{H}_{\mathrm{mac}}$ are uniform inside the sample.

Such an exception can arise only for ideal superconductors with their perfect diamagnetism. For other substances the requirement of a macroscopically local magnetic constitutive equation suffices to fix the natural gauge uniquely.

3. Effects of medium-carried explicit current density

In Sec. II.E it was noted that a modified formulation of electromagnetism in media in which medium-carried explicit densities (MCED'S) are included is sometimes useful. Here some further comments on the motivation and interpretation of the modified formulation will be made.

Although the formulation including medium-carried explicit densities can in principle be used in the dynamic regime, the present discussion will be restricted to the quasistatic regime, which motivates the key definitions

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[15] The divergence can be chosen arbitrarily inside the sample, but must be left free at the boundary surface if $\mathbf{M}_{\mathrm{mac}}$ is to vanish outside the sample, as is natural in a macroscopic description.

[16] Standard theorems from vector analysis guarantee the existence of an $\mathbf{M}_{\mathrm{mac}}(\mathbf{r})$ to generate an arbitrary steady state $\mathbf{J}_{\mathrm{mac}}(\mathbf{r})$ in an isolated simply connected sample. To describe $\mathbf{J}_{\mathrm{mac}}(\mathbf{r})$ in a multiply-connected sample, a medium-carried explicit current density may be necessary, as is the case for a circulating current in a superconducting ring.

and covers the applications of main interest. In a natural description **P** is then time independent and contributes no current density. It will be assumed that "foreign" current occurs only outside the medium, as holds for typical applications. Then the current density inside the medium is

$$\mathbf{J}=\mathbf{J}_{\mathrm{MCED}}+c\nabla\times\mathbf{M}. \tag{B3.1}$$

Since by Stokes's theorem the second term can yield no net flow across any cross section of the medium or into any closed part of it, $\mathbf{J}_{\mathrm{MCED}}$ is responsible for all charge transport. At first sight it might seem that a natural "division of labor" could be achieved by requiring the term in **M** to generate the entire curl of **J**, i.e., by postulating

$$\nabla\times\mathbf{J}_{\mathrm{MCED}}\equiv 0. \tag{B3.2}$$

However, further thought shows this not to be a viable choice: since in the quasistatic regime, we also have $\nabla\cdot\mathbf{J}_{\mathrm{MCED}}=0$, it would follow that $\mathbf{J}_{\mathrm{MCED}}$ could be fixed entirely by its boundary values, independent of the physical properties of the specific medium at hand, which would be artificial.

To allow a useful treatment of the relevant applications, $\mathbf{J}_{\mathrm{MCED}}$ must instead be defined by a suitable constitutive equation, which in the quasistatic case means a conduction law. Although the formulation so obtained, inevitably has a phenomenological aspect due to use of a macroscopic constitutive equation in a fundamental definition, it provides a natural physical description. One must, however, be on guard against interpretational pitfalls arising from the fact that the total magnetic dipole moment of the sample will, in general, contain contributions from $\mathbf{J}_{\mathrm{MCED}}$ and so can no longer be identified with the integral of $\mathbf{M}(\mathbf{r})$.

For example, the usual description of ferromagnetic resonance in a metal includes an explicit current density which obeys a conduction law and is to be identified with the present $\mathbf{J}_{\mathrm{MCED}}$. From explicit solutions (Kittel, 1948), it can be seen that the radio-frequency magnetic dipole moment contains contributions from $\mathbf{J}_{\mathrm{MCED}}$ as well as **M**, but only **M** is phenomenologically identifiable as a magnetization whose motion can be described by a gyromagnetic equation.

As a second example, consider a sample of uniform cross section through which a uniform static $\mathbf{J}_{\mathrm{MCED}}$ is flowing, being fed by foreign current at the end surfaces. The contribution of $\mathbf{J}_{\mathrm{MCED}}$ to the dipole moment of the sample is mathematically well defined, but nevertheless artificial, as shown by the fact that its value depends on the choice of origin. Such a contribution does not carry the usual physical significance of a static magnetic dipole moment; for example, it cannot be determined by conventional magnetometry methods because of forces on the current-carrying leads. Again only the contribution from **M** is phenomenologically identifiable as a magnetization.

4. Significance of the magnetic fields B and H

The distinction between the magnetic fields **B** and **H** has not always correctly been made in the literature. Upon closer examination, some of the confusion is found to be merely a matter of notation,[17] but in other cases genuine physics is at issue. Let us review the basic considerations that fix the roles played by **B** and **H**.

The auxiliary magnetic field **H** is defined in terms of the fundamental magnetic field **B** by

$$\mathbf{B}\equiv\mathbf{H}+4\pi\mathbf{M}. \tag{B4.1}$$

If **B** and **H** are separated into parts "med" and "ext" generated by the medium itself and by external sources, the term in **M** obviously must be associated with the former, so we have

$$\mathbf{B}_{\mathrm{med}}\equiv\mathbf{H}_{\mathrm{med}}+4\pi\mathbf{M}, \tag{B4.2}$$

$$\mathbf{B}_{\mathrm{ext}}\equiv\mathbf{H}_{\mathrm{ext}}. \tag{B4.3}$$

For an elongated sample oriented parallel to an external field, the geometry of most practical interest, demagnetization effects are large if expressed as corrections to **B**, but small as corrections to **H**. The standard choice of **H** rather than **B** as field variable in the macroscopic magnetic constitutive equation,

$$\mathbf{M}_{\mathrm{mac}}(\mathbf{r})=\mathbf{M}_{\mathrm{mac}}[\mathbf{H}_{\mathrm{mac}}(\mathbf{r})], \tag{B4.4}$$

is motivated by the wish to make demagnetization corrections small in such a geometry. Although $\mathbf{H}_{\mathrm{ext}}$ and $\mathbf{B}_{\mathrm{ext}}$ are both admissible as notations for an external magnetic field, $\mathbf{H}_{\mathrm{ext}}$ is usually written because it harmonizes better with the standard constitutive equation.

It is its appearance in the expression for the Lorentz force on a particle of charge q,

$$\mathbf{F}=q[\mathbf{E}+(\mathbf{v}/c)\times\mathbf{B}], \tag{B4.5}$$

which marks **B** as the fundamental magnetic field.[18] The auxiliary magnetic field **H** is useful for writing the magnetic constitutive equation (and for other "internal" calculations for a medium, as illustrated in Appendix B.5), but it can never naturally appear in an expression such as Eq. (B4.5) for an electromagnetic force exerted by the medium on a foreign probe.

To illustrate, let us recall the coupling between a medium and an external magnetic field obtained in Sec. III.A, Eq. (3.7),

$$H_{\mathrm{per}}=-\int d^3r\mathbf{M}(\mathbf{r},t)_T\cdot\mathbf{H}_{\mathrm{per}}(\mathbf{r},t).$$

In this expression $\mathbf{H}_{\mathrm{per}}$ denotes a magnetic field that is external and so could equally well have been called $\mathbf{B}_{\mathrm{per}}$. In the application to neutron scattering $\mathbf{H}_{\mathrm{per}}$ is the field generated by the neutron, which is external because the

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[17] An extremely unfortunate notation often used in the older literature, in which the macroscopic and microscopic **B** fields were written as **B** and **h**, rather than **B** and **b** as would have been logical, has been the source of much confusion.

[18] The electric fields **E** and **D** are the fundamental and auxiliary ones, respectively, for the same reason.

neutron is foreign to the medium. For neutron optics, on the other hand, the appropriate coupling is

$$H_{\mathrm{per}}=-(\mathbf{m}_{M1})_n\cdot\mathbf{B}(\mathbf{r}_n,t), \tag{B4.6}$$

where $(\mathbf{m}_{M1})_n$ is the magnetic dipole-moment operator of the neutron and $\mathbf{B}(\mathbf{r}_n,t)$ is the total **B** field at the neutron's position. In this case **B** and **H** are no longer equal because they are generated in part by the medium. In the 1930s and 40s there was a controversy as to whether the field in the coupling should be **H**, as maintained by Bloch (1936), or **B**, as maintained by Schwinger (1937). A very interesting account of this controversy can be found in Sears (1986). Here it suffices to note that Schwinger's result, to which the general considerations just reviewed would point, is indeed the correct one.[19]

5. Magnetic charge density

For a static medium in which the current density is generated entirely by **M**, it is sometimes convenient to define a magnetic charge density by

$$\rho_{\mathrm{mag}}(\mathbf{r})\equiv-\nabla\cdot\mathbf{M}(\mathbf{r}). \tag{B5.1}$$

The **H** field is then determined by the equations

$$\nabla\cdot\mathbf{H}=4\pi\rho_{\mathrm{mag}}, \tag{B5.2}$$

$$\nabla\times\mathbf{H}=0, \tag{B5.3}$$

and so can be found by methods familiar from electrostatics.

A practical application is the calculation of the macroscopic magnetization $\mathbf{M}_{\mathrm{mac}}(\mathbf{r})$ induced by a given external field $\mathbf{H}_{\mathrm{ext}}(\mathbf{r})$ in a sample of given shape that satisfies a known constitutive equation of the macroscopically local form

$$\mathbf{M}_{\mathrm{mac}}(\mathbf{r})=\mathbf{M}_{\mathrm{mac}}[\mathbf{H}_{\mathrm{mac}}(\mathbf{r})], \tag{B5.4}$$

where $\mathbf{H}_{\mathrm{mac}}\equiv\mathbf{H}_{\mathrm{ext}}+\mathbf{H}_{\mathrm{dem}}$ and $\mathbf{H}_{\mathrm{dem}}(\mathbf{r})$ is the demagnetization field. This problem can be solved by starting from the approximate solution

$$\mathbf{M}_{\mathrm{mac}}(\mathbf{r})=\mathbf{M}_{\mathrm{mac}}[\mathbf{H}_{\mathrm{ext}}(\mathbf{r})] \tag{B5.5}$$

and iterating numerically according to the scheme

$$\mathbf{M}_{\mathrm{mac}}(\mathbf{r})\Rightarrow\rho_{\mathrm{mag}}(\mathbf{r})\Rightarrow\mathbf{H}_{\mathrm{dem}}(\mathbf{r})\Rightarrow\mathbf{H}_{\mathrm{mac}}(\mathbf{r})\Rightarrow\mathbf{M}_{\mathrm{mac}}(\mathbf{r}) \tag{B5.6}$$

until a self-consistent solution is obtained. The use of the scalar field $\rho_{\mathrm{mag}}(\mathbf{r})$ rather than a current density to describe the state of the sample simplifies the calculation substantially.

The dependence of the magnetic charge density on the gauge of the magnetization (which in the above example is fixed by the constitutive equation) shows that there is no possibility of measuring its local value directly, not even macroscopically. It is remarkable that it should, nevertheless, be useful in such an eminently practical application as the one just sketched.

APPENDIX C: LINEAR RESPONSE AND SCATTERING—SUPPLEMENTARY DISCUSSION

1. Van Hove formulas for magnetic neutron scattering

Van Hove formulas show how cross sections for inelastic magnetic neutron scattering are related to autocorrelations of the magnetization described by the dynamic magnetic structure factor. For a full determination of this structure factor, cross sections for scattering of a polarized beam with polarization-sensitive detection of the scattered neutrons must be measured.

The spin state of a polarized beam of neutrons is described by a density matrix

$$\rho=(1/2)(\mathbb{1}+\mathbf{P}\cdot\boldsymbol{\sigma}). \tag{C1.1}$$

Here $\boldsymbol{\sigma}$ is the vector operator whose components are the Pauli matrices, and the polarization vector **P** is the expectation value of $\boldsymbol{\sigma}$ in the beam. The spin state of the neutrons after scattering is similarly described by

$$\rho'=(1/2)(\mathbb{1}+\mathbf{P}'\cdot\boldsymbol{\sigma}), \tag{C1.2}$$

where the polarization vector $\mathbf{P}'$ depends on the specific scatterer and on variables such as the scattering angle.

A polarization-sensitive detector detects such scattered neutrons with probability $\mathrm{Tr}\{\rho' D\}$, where D is a Hermitian positive-semidefinite matrix in the space of neutron-spin states. The most general such matrix is

$$D=d(\mathbb{1}+\mathbf{P}''\cdot\boldsymbol{\sigma}), \tag{C1.3}$$

where $D>0$ and $\mathbf{P}''$ is a polarization vector characterizing the detector. For an ideal detector, $d=1/2$ and $|\mathbf{P}''|=1$; then the scattered neutrons are detected with a probability

$$\mathrm{Tr}\{\rho' D\}=(1+\mathbf{P}'\cdot\mathbf{P}'')/2, \tag{C1.4}$$

which ranges from 1 for $\mathbf{P}'=\mathbf{P}''$ to zero for $\mathbf{P}'=-\mathbf{P}''$.

The cross section for inelastic magnetic neutron scatterin in the Born approximation, as observed by an ideal polarization-sensitive detector, is

$$\begin{aligned}(d^2\sigma/d\Omega\ dE')_{P''P}&=(k_f/k_i)(r_0{}^2/2)(1/2)\\&\quad\times\mathrm{Tr}[\langle(\mathbb{1}+\mathbf{P}\cdot\boldsymbol{\sigma})\mathbf{M}(\mathbf{k},\omega)_T\cdot\boldsymbol{\sigma}(\mathbb{1}+\mathbf{P}''\cdot\boldsymbol{\sigma})\mathbf{M}(\mathbf{k})_T^{\dagger}\cdot\boldsymbol{\sigma}\rangle],\end{aligned} \tag{C1.5}$$

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[19] The widespread belief that a model in which the neutron is a point dipole leads to a coupling proportional to **H**, while a model in which it is a current distribution leads to a coupling proportional to **B**, so that experiments showing the coupling to be proportional **B** demonstrate the correctness of the latter model, is patently fallacious in view of the fact that a magnetic dipole moment is nothing more than an abbreviated description of a current distribution. This misinterpretation is the result of symbol fetishism: customary but objectively meaningless choices between the equivalent symbols $\mathbf{B}_{\mathrm{ext}}$ and $\mathbf{H}_{\mathrm{ext}}$ in different types of calculations involving an external magnetic field have been carried over to the coupling to the magnetic field in a medium, where they are invalid.

where $\hbar\mathbf{k}_i$ and $\hbar\mathbf{k}_f$ are the initial and final momenta of the neutron, $\mathbf{k}\equiv(\mathbf{k}_i-\mathbf{k}_f)$ is the momentum transfer, $\omega \equiv(k_i^2-k_f^2)\hbar^2/2m_n$ is the energy transfer, $\mathbf{M}(\mathbf{k})_T$ is a Schrödinger operator, $\mathbf{M}(\mathbf{k},\omega)_T$ is the Fourier transform of a Heisenberg operator, $r_0\equiv g_n e^2/m_e c^2$ is the magnetic scattering length, with g_n the gyromagnetic ratio of the neutron, and $\langle\,\rangle$ is a thermal average over states of the sample. Evaluation of the trace over neutron spin states yields

$$\begin{aligned}(1/2)\ \mathrm{Tr}\{\ \}\equiv&\langle\mathbf{M}(\mathbf{k},\omega)_T\cdot\mathbf{M}(\mathbf{k})_T^\dagger\rangle(1-\mathbf{P}''\cdot\mathbf{P})\\&+i(\mathbf{P}-\mathbf{P}'')\cdot\langle\mathbf{M}(\mathbf{k},\omega)_T\times\mathbf{M}(\mathbf{k})_T^\dagger\rangle\\&+\mathbf{P}\cdot\langle\mathbf{M}(\mathbf{k},\omega)_T\mathbf{M}(\mathbf{k})_T^\dagger\rangle\cdot\mathbf{P}''\\&+\mathbf{P}''\cdot\langle\mathbf{M}(\mathbf{k},\omega)_T\mathbf{M}(\mathbf{k})_T^\dagger\rangle\cdot\mathbf{P}.\end{aligned}\tag{C1.6}$$

The dynamic magnetic structure factor[20] is the 3×3 Hermitian matrix

$$S(\mathbf{k},\omega)_{ii'}\equiv\langle M_T(\mathbf{k},\omega)_i(M_T^\dagger(\mathbf{k})_{i'}\rangle,\tag{C1.7}$$

which is also a second-rank Cartesian tensor. As expressed in terms of this tensor, the cross section of Eq. (C1.5) is

$$\begin{aligned}&(d^2\sigma/d\Omega\ dE')_{P''P}\\&=(k_fK_i)(r_0^2)\Bigg\{(1/2)(1-\mathbf{P}''\cdot\mathbf{P})\sum_j S(\mathbf{k},\omega)_{jj}\\&\quad+(1/2)i\sum_{jj'j''}\varepsilon_{j\,j'j''}(P_j-P_j'')S(\mathbf{k},\omega)_{j'j''}\\&\quad+(1/2)\sum_{jj'}P_{j'}[S(\mathbf{k},\omega)_{j\,j'}+S(\mathbf{k},\omega)_{j'j}]\Bigg\},\end{aligned}\tag{C1.8}$$

where $\varepsilon_{j\,j'j''}$ is the totally antisymmetric Levi-Civita tensor density. In this expression $|\mathbf{P}''|=1$, but $|\mathbf{P}|\leqslant1$ is allowed, and the cross section with an unpolarized beam, but polarization-sensitive detection is just that obtained by inserting $\mathbf{P}=\mathbf{0}$:

$$\begin{aligned}(d^2\sigma/d\Omega dE')_{P''0}=(k_f/k_i)(r_0^2)\Bigg\{&(1/2)\sum_j S(\mathbf{k},\omega)_{jj}\\&-(1/2)i\sum_{jj'j''}\varepsilon_{jj'j''}P_j''S(\mathbf{k},\omega)_{j'j''}\Bigg\}.\end{aligned}\tag{C1.9}$$

Results for polarization-independent detection are obtained by summing $\mathbf{P}''$ over $\pm\mathbf{u}$, where $\mathbf{u}$ is an arbitrary unit vector. The cross section for a polarized beam with unpolarized detection is thus

$$\begin{aligned}(d^2\sigma/d\Omega dE')_P=(k_f/k_i)(r_0^2)\Bigg\{&\sum_j S(\mathbf{k},\omega)_{jj}\\&+i\sum_{jj'j''}\varepsilon_{jj'j''}P_jS(\mathbf{k},\omega)_{j'j''}\Bigg\},\end{aligned}\tag{C1.10}$$

and the cross section for an unpolarized beam with unpolarized detection is

$$(d^2\sigma/d\Omega dE')_{\mathrm{unpol}}=(k_f/k_i)(r_0^2)\sum_j S(\mathbf{k},\omega)_{j\,j}.\tag{C1.11}$$

The scattering cross section for an unpolarized beam and unpolarized detection thus determines only the trace of the structure factor, which is its part of spherical tensor rank 0. The cross section for an unpolarized beam and polarized detector or a polarized beam and unpolarized detector determines in addition the antisymmetric part of the matrix $S(\mathbf{k},\omega)_{i\,i'}$, which is also its imaginary part and the part of spherical tensor rank 1. To determine the remaining part, the trace-free symmetrized part, which is the part of spherical tensor rank 2, cross sections for a polarized beam with a polarized detector must be measured. The nine cross sections defined by allowing each of the polarization vectors $\mathbf{P}$ and $\mathbf{P}''$ to range over three linearly independent values contain information that is just sufficient to determine the entire tensor $S(\mathbf{k},\omega)_{ii'}$ for given $\mathbf{k}$ and ω.

2. Fluctuations, dissipation, and scattering

The fundamental relations that connect the spontaneous fluctuations in an unperturbed system, the dissipative part of the dynamic susceptibility describing its linear response to a classical external field, and the autocorrelation function implying inelastic scattering cross sections, are described in many reviews, of which Callen (1962) and Kubo (1966) may especially be mentioned. These relations apply in their usual forms to the magnetization operator $\mathbf{M}(\mathbf{r},t)$ as defined in this articl, so no further discussion of the fluctuation concepts *per se* is necessary. However, there are two rather technical points for which clarification may be in order. The first is the distinction between the two-point magnetic susceptibility needed to describe a solid at the microscopic level and the one-point reduced susceptibility to which the scattering can be related. The second is the remark that it is the anti-Hermitian part of the susceptibility matrix which fixes the dissipation, not the imaginary part as is sometimes implied in the literature.

Unless a continuum is homogeneous, a two-point susceptibility tensor is needed to describe it. Since no real solid is homogeneous on the atomic scale, the susceptibility tensor which describes it in the first instance is a two-point one. It is defined in the $(\mathbf{r},t)$ representation by

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[20] In this article, all Fourier transforms $f(t)\rightarrow f(\omega)$ are defined with the normalization used in Eq. (3.10), which gives the relations from linear-response theory summarized in Appendix C.2 their most natural forms. The present $S(\mathbf{k},\omega)_{ii'}$ is consequently larger by a factor 2π than that defined by Marshall and Lovesey (1971).

$$M_T(\mathbf{r},t)_i=\sum_{i'}\int d^3r'\int dt'\chi(\mathbf{r},\mathbf{r}';t-t')_{ii'}H(\mathbf{r}',t')_{i'} \tag{C2.1}$$

where **M** is the deviation of the magnetization from its value at equilibrium and i is a Cartesian component. After transformation to the **k** representation [with $M_T(t)_{i\mathbf{k}}\equiv M_T(\mathbf{k},t)_i$, etc.] this becomes

$$M_T(t)_{i\mathbf{k}}=\sum_{i'\mathbf{k}'}\int dt'\chi(t-t')_{i\mathbf{k};i'\mathbf{k}'}H(t')_{i'\mathbf{k}'}. \tag{C2.2}$$

The two-point susceptibility appearing in this expression is what is given by the Kubo formula, Eq. (3.8).

The rate of work done on the continuum by **H** is

$$dW/dt=\sum_{i\mathbf{k}}\mathbf{H}(t)_{i\mathbf{k}}(d/dt)\mathbf{M}_T(t)_{i\mathbf{k}}. \tag{C2.3}$$

By substituting Eq. (C2.2), integrating over time, and converting to the ω representation, one finds the work done by a field pulse of limited duration to be

$$W=\sum_{i\mathbf{k}i'\mathbf{k}'}\int(d\omega/2\pi)\omega[\mathscr{Ah}\chi(\omega)]_{i\mathbf{k};i'\mathbf{k}'}\times H(\omega)^*_{i\mathbf{k}}H(\omega)_{i'\mathbf{k}'}. \tag{C2.4}$$

A general matrix A can be decomposed into Hermitian and anti-Hermitian parts, $\mathscr{He}A$ and $\mathscr{Ah}A$, by

$$A\equiv(A+A^\dagger)/2+i[(A-A^\dagger)/2i]\equiv\mathscr{He}A+i\mathscr{Ah}A, \tag{C2.5}$$

and what appears in Eq. (C2.4) is the anti-Hermitian part of χ, read as a matrix in the compound index $i\mathbf{k}$. The anti-Hermitian part of χ is thus associated with dissipation, and the remaining Hermitian part is associated with dispersion. The same Kramers-Kronig relations that are usually given for the real and imaginary parts of χ apply equally well for $\mathscr{He}\chi$ and $\mathscr{Ah}\chi$, allowing either to be recovered from the other.

Standard manipulations starting from the Kubo formula allow the anti-Hermitian part of the susceptibility to be expressed as

$$[\mathscr{Ah}\chi(\omega)]_{i\mathbf{k};i'\mathbf{k}'}=(1/2)(1-e^{-\beta\omega})\times\langle\mathbf{M}_T(\omega)_{i\mathbf{k}}(\mathbf{M}_T^\dagger)_{i'\mathbf{k}'}\rangle, \tag{C2.6}$$

where $\mathbf{M}_T(\omega)_{i\mathbf{k}}$ is the Fourier transform of $\mathbf{M}_T(t)_{i\mathbf{k}}$. Canonical fluctuations are defined by

$$F(t)_{i\mathbf{k};i'\mathbf{k}'}\equiv(1/2)\langle\mathbf{M}_T(t)_{i\mathbf{k}}(\mathbf{M}_T^\dagger)_{i'\mathbf{k}'}+(\mathbf{M}_T^\dagger)_{i'\mathbf{k}'}\mathbf{M}_T(t)_{i\mathbf{k}}\rangle, \tag{C2.7}$$

and the relation between their frequency spectrum and the dissipative part of the susceptibility (the fluctuation-dissipation theorem) is

$$F(\omega)_{i\mathbf{k};i'\mathbf{k}'}=\coth(\beta\omega/2)[\mathscr{Ah}\chi(\omega)]_{i\mathbf{k};i'\mathbf{k}'}. \tag{C2.8}$$

What can be determined by magnetic neutron scattering is only the dynamic magnetic structure factor, which by (C2.6) corresponds to the part of the susceptibility with $\mathbf{k}=\mathbf{k}'$, i.e., a reduced one-point susceptibility defined by

$$\chi_{\text{red}}(\mathbf{k};\omega)_{ii'}\equiv\chi(\omega)_{i\mathbf{k};i'\mathbf{k}} \tag{C2.9}$$

or equivalently in the **r** representation by

$$\chi_{\text{red}}(\mathbf{r};\omega)_{ii'}\equiv(1/V_{\text{cell}})\int d^3r'\chi(\mathbf{r}+\mathbf{r}',\mathbf{r}';\omega)_{ii'}. \tag{C2.10}$$

Combining these expressions, one finds the dynamic magnetic structure factor to be proportional to the anti-Hermitian part of the reduced susceptibility tensor as indicated in Eq. (3.12).

APPENDIX D: THE MICROSCOPIC MAGNETIZATION—SUPPLEMENTARY TOPICS

1. Spin magnetization and spin density

The theoretically obvious description of spin magnetization is

$$\mathbf{M}_{\text{spin}}(\mathbf{r})=g_0\mu_A\mathbf{s}(\mathbf{r}), \tag{D1.1}$$

where $\mathbf{s}(\mathbf{r})$ is the density of electron-spin angular momentum, g_0 is the gyromagnetic factor of the free electron, $\mu_A\equiv-\mu_B$ is the "anti-Bohr" magneton,[21] and the product on the right-hand side is the dipole-moment density due to electron spin.

Actually any magnetization can be identified as a density of *effective* magnetic dipole moments by a formula similar to that above. This means simply that such a density $\mathbf{M}(\mathbf{r})$ would generate the correct transverse current density by the formula $\mathbf{J}(\mathbf{r})_T=c\nabla\times\mathbf{M}(\mathbf{r})$, and it is more a verbal convention than a physical description so long as the effective moments have no objective existence, as in the case of orbital magnetization.

Spin magnetization is exceptional in that underlying magnetic dipole moments do exist objectively. However, although their density is in principle observable, it cannot be determined by purely electromagnetic measurements. The size of an isolated dipole moment can, of course, be measured electromagnetically, but the density of a distribution of such moments cannot be so determined because the contributions of the individual moments cannot be distinguished. Any observation of the spin density must refer back to its definition as a spin-weighted particle density, e.g., by a hypothetical experiment in which electrons are knocked out of the sample and counted by a spin-sensitive particle detector. Because of the use of a particle detector, such a measurement would not be purely electromagnetic.[22]

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[21] Awkward minus signs and confusion as to the meaning of "spin up" can be avoided by writing all equations in terms of μ_A and the electron charge $e_e\equiv-|e|$. When expressed in units of μ_A, spin and orbital magnetic moments are parallel rather than confusingly antiparallel to the corresponding angular momenta.

[22] Magnetic neutron scattering is a purely electromagnetic measurement, despite the use of a particle detector, because the neutrons that are detected belong to the apparatus, not to the system being observed.

Since the spin magnetization belongs to electromagnetic theory, it would be illogical and injudicious to define it in terms of the spin density, which cannot be measured electromagnetically. Although the "spin-density gauge" defined by Eq. (D1.1) is the most natural description when the spin density is known, it should be regarded as only one possible gauge choice and not as a universally valid definition of the spin magnetization. In other words, the spin magnetization should be defined only in terms of the spin-carried electric current density. The appropriateness of this definition is confirmed by the fact that the electromagnetic measurements discussed in Sec. III all "see" spin magnetization only as a current density.

2. Orbital magnetization and angular momentum

As seen in Appendix D.1, the spin magnetization is simply related to the spin density (the density of spin angular momentum) provided that it is expressed in a suitable gauge. The gauge in which the orbital magnetization is most simply related to the density of orbital angular momentum is the Trammell gauge. Following Balcar (1975), let us display this relation for the orbital magnetization $\langle \mathbf{m}_{\mathrm{orb}}(\mathbf{r})\rangle$ corresponding to a localized atomic or cellular orbital current density $\langle \mathbf{j}_{\mathrm{orb}}(\mathbf{r})\rangle$, to which the Trammell gauge applies most immediately. (All quantities in the following are understood to be purely orbital, but the subscript "orb" will usually be omitted.)

The electric current operator for electron n is

$$\mathbf{I}_n \equiv (e_e/m_e)\mathbf{p}_n . \tag{D2.1}$$

The operator for the electric current density at field point $\mathbf{r}$ is

$$\mathbf{j}(\mathbf{r}) \equiv \sum_n (e_e/2m_e)[\mathbf{p}_n \delta(\mathbf{r}_n - \mathbf{r}) + \delta(\mathbf{r}_n - \mathbf{r})\mathbf{p}_n]. \tag{D2.2}$$

The operator for the angular momentum of electron n (in units $\hbar$) is

$$\mathbf{l}_n \equiv \mathbf{r}_n \times \mathbf{p}_n/\hbar. \tag{D2.3}$$

The density operator for angular momentum is

$$\mathbf{l}(\mathbf{r}) \equiv \sum_n (1/2)[\mathbf{l}_n \delta(\mathbf{r}_n - \mathbf{r}) + \delta(\mathbf{r}_n - \mathbf{r})\mathbf{l}_n]. \tag{D2.4}$$

This density operator is related to the current-density operator by

$$\mathbf{r} \times \mathbf{j}(\mathbf{r}) \equiv (e_e \hbar/m_e)\mathbf{l}(\mathbf{r}). \tag{D2.5}$$

The orbital magnetization in the Trammell gauge at thermal equilibrium can thus be expressed in terms of the expectation value of the density operator for orbital angular momentum as

$$\begin{aligned}\langle \mathbf{m}_{\mathrm{orb}}(\mathbf{r})\rangle &\equiv (e_e\hbar/m_e c)\int_1^\infty d\lambda \langle \mathbf{l}(\lambda \mathbf{r})\rangle \\ &\equiv 2\mu_A \int_1^\infty d\lambda \langle \mathbf{l}(\lambda \mathbf{r})\rangle.\end{aligned} \tag{D2.6}$$

It should be noted that, although $\mu_A\langle \mathbf{l}(\mathbf{r})\rangle$ integrates to $\mu_A\langle \mathbf{l}\rangle$, which is the correct atomic orbital magnetic dipole moment, it is not itself an acceptable atomic magnetization because

$$c\nabla \times [\mu_A \langle \mathbf{l}(\mathbf{r})\rangle] \not\equiv \langle \mathbf{j}_{\mathrm{orb}}(\mathbf{r})\rangle. \tag{D2.7}$$

3. Atomic or cellular current density

Let $\langle \mathbf{j}(\mathbf{r})\rangle$ be the electric current density in an atom or cell centered at the origin. When $\langle \mathbf{j}(\mathbf{r})\rangle$ is exponentially bounded at large $\mathbf{r}$, as is usually the case, its Fourier transform $\langle \mathbf{j}(\mathbf{k})\rangle$ is analytic for real $\mathbf{k}$. Let us examine the first terms in its expansion in powers of the components of $\mathbf{k}$.

Partial integration of the steady-flow condition

$$\nabla \cdot \langle \mathbf{j}(\mathbf{r})\rangle = 0 \tag{D3.1}$$

shows that the constant term $\langle \mathbf{j}(\mathbf{k}{=}0)\rangle$ vanishes. The general expansion to linear order is

$$\langle \mathbf{j}(\mathbf{k})\rangle_j \equiv \sum_{j'} iC_{jj'}\mathbf{k}_{j'}, \tag{D3.2}$$

where the reality of $\langle \mathbf{j}(\mathbf{r})\rangle$ implies reality of $C_{jj'}$. The matrix C can be decomposed into real symmetric and real antisymmetric parts, and the former must vanish as follows from $\mathbf{k}\cdot\langle \mathbf{j}(\mathbf{k})\rangle = 0$ after a principal-axis transformation. The remaining antisymmetric part can be written as $ic\mathbf{k}\times\langle \mathbf{m}_{M1}\rangle$, where $\mathbf{m}_{M1}$ is the magnetic dipole moment operator for the current distribution:

$$\mathbf{m}_{M1} \equiv (1/2c)\int d^3r\ \mathbf{r}\times\mathbf{j}(\mathbf{r}). \tag{D3.3}$$

The expansion of $\langle \mathbf{j}(\mathbf{k})\rangle$ is thus

$$\langle \mathbf{j}(\mathbf{k})\rangle = ic\mathbf{k}\times\langle \mathbf{m}_{M1}\rangle + O(k^2). \tag{D3.4}$$

The transverse part of the magnetization is

$$\langle m(\mathbf{k})_T\rangle \equiv (i/c)\mathbf{k}\times\langle \mathbf{j}(\mathbf{k})\rangle/\mathbf{k}\cdot\mathbf{k} \tag{D3.5}$$

and its expansion is

$$\langle \mathbf{m}(\mathbf{k})_T\rangle = (\mathbf{k}\times\langle \mathbf{m}_{M1}\rangle)\times\mathbf{k}/\mathbf{k}\cdot\mathbf{k} + O(k^1), \tag{D3.6}$$

where the leading term is the point-dipolar or small-κ approximation.

If a gauge such as the Trammell gauge is chosen which yields a well-bounded $\langle \mathbf{m}(\mathbf{r})\rangle$ at large $\mathbf{r}$, then $\langle \mathbf{m}(\mathbf{k})\rangle$ will be analytic at small k, and from Eq. (D3.6) its expansion is

$$\langle \mathbf{m}(\mathbf{k})\rangle = \langle \mathbf{m}_{M1}\rangle + O(k^1). \tag{D3.7}$$

In the $\mathbf{r}$ representation the point-dipolar approximation is then

$$\langle \mathbf{m}(\mathbf{r})\rangle = \langle \mathbf{m}_{M1}\rangle\delta(\mathbf{r}), \tag{D3.8}$$

showing that it indeed describes a point dipole.

Instead of interpolating $\langle \mathbf{J}(\mathbf{K})\rangle$ to obtain the cellular current density $\langle \mathbf{j}(\mathbf{k})\rangle$ and hence $\langle \mathbf{m}_{M1}\rangle$, as contemplated in Sec. IV.A, one can obtain $\langle \mathbf{m}_{M1}\rangle$ more directly by extrapolating $\langle \mathbf{M}(\mathbf{K}_\perp)_T\rangle$ to $\mathbf{K}_\perp = 0$. Although $\langle \mathbf{M}(\mathbf{K}_\perp)_T\rangle$ and $\langle \mathbf{M}(\mathbf{K}_\perp)\rangle$ are equal at finite $\mathbf{K}_\perp$ only if $\langle \mathbf{M}(\mathbf{K})\rangle$ is perfectly parallel, from Eq. (D3.6) it follows that their values in the limit $\mathbf{K}_\perp=0$ are always equal if $\mathbf{u}_\parallel$ is taken as the direction of $\langle \mathbf{M}_{\mathbf{M1}}\rangle$.

4. The Trammell gauge—formal solutions

The definition of an atomic magnetization in the Trammell gauge can be written for thermal expectation values

$$\langle \mathbf{m}(\mathbf{r})\rangle \equiv (1/c)\int_1^\infty d\lambda\,\lambda\mathbf{r}\times\langle \mathbf{j}(\lambda\mathbf{r})\rangle \tag{D4.1}$$

or more generally as the dynamically valid operator relation

$$\mathbf{m}(\mathbf{r})\equiv(1/c)\int_1^\infty d\lambda\,\lambda\mathbf{r}\times\mathbf{j}(\lambda\mathbf{r})_T. \tag{D4.2}$$

That the latter expression indeed satisfies $c\nabla\times\mathbf{m}(\mathbf{r})=\mathbf{j}(\mathbf{r})_T$ can be demonstrated with the help of the lemma

$$\lambda(d/d\lambda)f(\lambda\mathbf{r})=\mathbf{r}\cdot\nabla f(\lambda\mathbf{r}), \tag{D4.3}$$

which follows from the chain rule for differentiation.

Straightforward transformations show that the Trammell magnetization is given in the **k** representation by

$$\mathbf{m}(\mathbf{k})=(i/c)\nabla_{\mathbf{k}}\times\int_1^\infty d\lambda\,\lambda^{-2}\mathbf{j}(\mathbf{k}/\lambda)_T. \tag{D4.4}$$

This equation shows that the determination of $\langle\mathbf{M}(\mathbf{K})\rangle$ by $\langle\mathbf{J}(\mathbf{K})\rangle$ in the crystalline Trammell gauge is nonlocal in **K**. This follows because $\langle\mathbf{M}(\mathbf{K})\rangle$ equals $\langle\mathbf{m}(\mathbf{K})\rangle$, which depends on values of $\langle\mathbf{j}(\mathbf{K}/\lambda)\rangle$ for all λ, which are obtained by an interpolation fixed by the values of $\langle\mathbf{J}(\mathbf{K}')\rangle$ at various $\mathbf{K}'$.

5. The most nearly collinear gauge—formal solutions

a. Solution for a crystal directly

In Sec. IV.B.2 it was shown that the requirement that the noncollinear part of the magnetization in a crystal be minimal, which defines the MNC gauge, yields the condition

$$\mathbf{K}_\perp\cdot\langle\mathbf{M}(\mathbf{K})\rangle_\perp=0\forall\mathbf{K}.$$

This implies the further relations

$$(\mathbf{K}_\perp\times\langle\mathbf{M}(\mathbf{K})\rangle_\perp)\times\mathbf{K}_\perp=\langle\mathbf{M}(\mathbf{K})\rangle_\perp\mathbf{K}_\perp\cdot\mathbf{K}_\perp\,, \tag{D5.1}$$

$$(\mathbf{K}_\perp\times\langle\mathbf{M}(\mathbf{K})\rangle_\parallel)\times\mathbf{K}_\perp=\langle\mathbf{M}(\mathbf{K})\rangle_\parallel\mathbf{K}_\perp\cdot\mathbf{K}_\perp\,, \tag{D5.2}$$

$$(\mathbf{K}_\parallel\times\langle\mathbf{M}(\mathbf{K})\rangle_\perp)\times\mathbf{K}_\perp=0. \tag{D5.3}$$

Combining these with $ic\mathbf{K}\times\langle\mathbf{M}(\mathbf{K})\rangle=\langle\mathbf{J}(\mathbf{K})\rangle$ yields

$$\langle\mathbf{M}(\mathbf{K}))\rangle_\perp\mathbf{K}_\perp\cdot\mathbf{K}_\perp=(i/c)\mathbf{K}_\perp\times\langle\mathbf{J}(\mathbf{K})\rangle_\parallel\,, \tag{D5.4}$$

$$\langle\mathbf{M}(\mathbf{K}))\rangle_\parallel\mathbf{K}_\perp\cdot\mathbf{K}_\perp=(i/c)\mathbf{K}_\perp\times\langle\mathbf{J}(\mathbf{K})\rangle_\perp\,, \tag{D5.5}$$

and consequently

$$\langle\mathbf{M}(\mathbf{K})\rangle=(i/c)\mathbf{K}_\perp\times\langle\mathbf{J}(\mathbf{K})\rangle/\mathbf{K}_\perp\cdot\mathbf{K}_\perp\quad(\mathbf{K}_\perp\neq0), \tag{D5.6}$$

$$\langle\mathbf{M}(\mathbf{K})\rangle=(\mathbf{K}\times\langle\mathbf{M}(\mathbf{K})_T\rangle)\times\mathbf{K}_\perp/\mathbf{K}_\perp\cdot\mathbf{K}_\perp\quad(\mathbf{K}_\perp\neq0). \tag{D5.7}$$

The values of $\langle\mathbf{M}(\mathbf{K})\rangle_\parallel$ at points where $\mathbf{K}_\perp=0$ are indeterminate because these values contribute nothing to $\langle\mathbf{J}(\mathbf{K})\rangle=ic\mathbf{K}\times\langle\mathbf{M}(\mathbf{K})\rangle$. In principle arbitrary values can be inserted, but to preclude artificial choices it seems better to give a definite prescription, namely, the "poor man's interpolation" defined by the average of values of $\langle\mathbf{M}(\mathbf{K})\rangle_\parallel$ at nearest-neighbor $\mathbf{K}'$, excluding $\mathbf{K}'$ for which $\mathbf{K}'_\perp$ also vanishes. For the value at $\mathbf{K}=0$, which is of more interest since it corresponds to the mean magnetization, a better interpolation of the type discussed in Appendix D.3 should be made.

For closer analysis of magnetic Bragg scattering, it is helpful to decompose the $\perp$ subspace into parts along a unit vector **v** parallel to $\mathbf{K}_\perp$ and a unit vector $\mathbf{w}\equiv\mathbf{u}_\parallel\times\mathbf{v}$. The generic $\langle\mathbf{M}(\mathbf{K})_\mathbf{T}\rangle$ has finite components along all three directions $\parallel$, **v**, and **w**, but the first two are not independent since

$$\mathbf{K}\cdot\langle\mathbf{M}(\mathbf{K})_T\rangle\equiv\mathbf{K}_\parallel\cdot\langle\mathbf{M}(\mathbf{K})_T\rangle_\parallel+\mathbf{K}_\perp\cdot\langle\mathbf{M}(\mathbf{K})_T\rangle_V\equiv0. \tag{D5.8}$$

This relation allows $\langle\mathbf{M}(\mathbf{K})_T\rangle_V$ to be eliminated and Eq. (D5.7) to be brought to the form displayed as Eq. (4.20) in the main text.

b. Solution by cellular construction

The solution for the cellular (or atomic) magnetization in the **k** representation has the same form as that given above except that the discrete **K**'s are replaced by a continuous **k**. Since the MNC solution (unlike that in the Trammell gauge) is local in **k**, solving for $\langle\mathbf{M}(\mathbf{K})\rangle$ in the crystal by interpolating $\langle\mathbf{J}(\mathbf{K})\rangle$ to find a cellular $\langle\mathbf{j}(\mathbf{k})\rangle$ and using the latter to calculate the cellular $\langle m(\mathbf{k})\rangle$ would yield the same solution as found for the crystal directly.[23]

c. Solution in the r representation

The MNC $\langle\mathbf{m}(\mathbf{r})\rangle$ due to an atomic or cellular current density can usually best be evaluated in the **r** representation. If the magnetization in the MNC gauge is $\langle\mathbf{m}(\mathbf{r})\rangle$, in a general gauge it is

$$\langle\mathbf{m}'(\mathbf{r})\rangle\equiv\langle\mathbf{m}(\mathbf{r})\rangle+\nabla g(\mathbf{r}), \tag{D5.9}$$

where $g(\mathbf{r})$ is arbitrary. The conditions determining the MNP $\langle\mathbf{m}(\mathbf{r})\rangle$ in the **r** representation are

$$\int d^3r|\langle\mathbf{m}(\mathbf{r})\rangle_\perp+\nabla_\perp g(\mathbf{r})|^2=\text{minimum} \tag{D5.10}$$

and

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[23] This statement applies for generic **K**, but not for points where $\mathbf{K}_\perp=0$, where the solution for the crystal directly is indeterminate. In the cellular solution the corresponding singularity at $\mathbf{k}_\perp=\mathbf{0}$ is handled by the usual prescriptions for Fourier integrals, which yield the same uniquely defined $\langle\mathbf{m}(\mathbf{r})\rangle$ as would also be obtained by solving directly in the **r** representation.

$$c\nabla\times\langle\mathbf{m}(\mathbf{r})\rangle=\langle\mathbf{j}(\mathbf{r})\rangle \tag{D5.11}$$

with the boundary condition $\langle\mathbf{m}(\mathbf{r})\rangle\rightarrow 0$ as $\mathbf{r}\rightarrow\infty$. Variation of $g(\mathbf{r})$ with partial integration shows that Eq. (D5.10) is equivalent to

$$\nabla_\perp\cdot\langle\mathbf{m}(\mathbf{r})\rangle=0. \tag{D5.12}$$

Equations (D5.11) and (D5.12) can be integrated to yield

$$\langle\mathbf{m}(\mathbf{r}_\perp,\mathbf{r}_\parallel)\rangle=(-1/2\pi c)\int d^2r'_\perp\,\frac{(\mathbf{r}_\perp-\mathbf{r}'_\perp)}{|\mathbf{r}_\perp-\mathbf{r}'_\perp|^2}\times\langle\mathbf{j}(\mathbf{r}'_\perp,\mathbf{r}_\parallel)\rangle. \tag{D5.13}$$

This solution shows that in the MNC gauge $\langle\mathbf{m}(\mathbf{r})\rangle$ is fixed by $\langle\mathbf{j}(\mathbf{r})\rangle$ in each plane separately. It can be used to find the magnetization from atomic current densities $\langle\mathbf{j}(\mathbf{r})\rangle$ calculated by cellular methods in band theory.

For $\langle\mathbf{j}(\mathbf{r})\rangle$ from shell-model states the solution can be carried further analytically. Cylindrical coordinates (ρ, ϕ, z) are appropriate, with $z\equiv z_\parallel$, and $\langle\mathbf{j}(\mathbf{r})\rangle$ is assumed to have the form

$$\langle\mathbf{j}(\mathbf{r})\rangle\equiv\langle\overline{\mathbf{j}}(\mathbf{r})\rangle+\langle\tilde{\mathbf{j}}(\mathbf{r})\rangle, \tag{D5.14}$$

$$\langle\overline{\mathbf{j}}(\mathbf{r})\rangle\equiv u_\phi\langle\overline{\mathbf{j}}(\rho,z)_\phi\rangle, \tag{D5.15}$$

$$\langle\tilde{\mathbf{j}}(\mathbf{r})\rangle\equiv\mathbf{u}_\rho\langle\tilde{j}(\rho,z)_\rho\rangle\,\sin(\nu\phi)+\mathbf{u}_\phi\langle\tilde{j}(\rho,z)_\phi\rangle\,\sin(\nu\phi)$$
$$+\mathbf{u}_z\langle\tilde{j}(\rho,z)_z\rangle\,\cos(\nu\phi). \tag{D5.16}$$

Here the nonoscillating term $\langle\overline{\mathbf{j}}(\mathbf{r})\rangle$ occurs for a magnetic ion in free space, and an additional oscillating term $\langle\tilde{\mathbf{j}}(\mathbf{r})\rangle$ of the indicated form is what typically results from a crystal field with ν-fold rotational symmetry about $\mathbf{u}_z$. The linearity of the differential equations allows one to solve separately for the corresponding parts $\langle\overline{\mathbf{m}}(\mathbf{r})\rangle$ and $\langle\tilde{\mathbf{m}}(\mathbf{r})\rangle$ of the magnetization.

Since the nonoscillating current $\langle\overline{\mathbf{j}}(\mathbf{r})\rangle$ is entirely perpendicular to $\mathbf{u}_z$, it generates a perfectly collinear magnetization

$$\langle\overline{\mathbf{m}}(\mathbf{r})\rangle=\mathbf{u}_z\langle\overline{\mathbf{m}}(\rho,z)_z\rangle. \tag{D5.17}$$

Equation (D5.12) is satisfied trivially and Eq. (D5.11) yields

$$\langle\overline{\mathbf{m}}(\rho,z)_z\rangle=(1/c)\int_\rho^\infty d\rho'\langle\overline{j}(\rho',z)_\phi\rangle. \tag{D5.18}$$

To solve for the oscillating part of the magnetization, $\langle\tilde{\mathbf{m}}(\mathbf{r})\rangle$, we seek a solution of the form

$$\langle\tilde{\mathbf{m}}(\mathbf{r})\rangle=\mathbf{u}_\rho\langle\tilde{\mathbf{m}}(\rho,z)_\rho\rangle\,\cos(\nu\phi)$$
$$+\mathbf{u}_\phi\langle\tilde{\mathbf{m}}(\rho,z)_\phi\rangle\,\sin(\nu\phi)$$
$$+\mathbf{u}_z\langle\tilde{\mathbf{m}}(\rho,z)_z\rangle\,\cos(\nu\phi). \tag{D5.19}$$

From Eq. (D5.12) it then follows that

$$\nu\langle\tilde{\mathbf{m}}(\rho,z)_\phi\rangle=-(\partial/\partial\rho)(\rho\langle\tilde{\mathbf{m}}(\rho,z)_\rho\rangle). \tag{D5.20}$$

We further assume $\mathbf{u}_r\cdot\langle\tilde{\mathbf{j}}(\mathbf{r})\rangle=0$, as holds for the current generated by standard shell-model states. As expressed in terms of cylindrical components, this condition reads

$$\langle\tilde{j}(\rho,z)_\rho\rangle=-(z/\rho)\langle\tilde{j}(\rho,z)_z\rangle. \tag{D5.21}$$

From this condition and the requirement $\nabla\cdot\langle\tilde{\mathbf{j}}(\mathbf{r})\rangle=0$, it follows that

$$\nu\langle\tilde{j}(\rho,z)_\phi\rangle=[\rho(\partial/\partial z)-z(\partial/\partial\rho)]\langle\tilde{j}(\rho,z)_z\rangle. \tag{D5.22}$$

The above four equations allow Eq. (D5.11) to be reduced to

$$\nu^2\langle\tilde{\mathbf{m}}(\rho,z)_\rho\rangle-(\nu\rho/c)\langle\tilde{j}(\rho,z)_z\rangle$$
$$=(\partial/\partial\rho)[\rho(\partial/\partial\rho)(\rho\langle\tilde{\mathbf{m}}(\rho,z)_\rho\rangle)] \tag{D5.23}$$

and

$$\nu^2\langle\tilde{\mathbf{m}}(\rho,z)_z\rangle-(\nu z/c)\langle\tilde{j}(\rho,z)_z\rangle$$
$$=(\partial/\partial z)[\rho(\partial/\partial\rho)(\rho\langle\tilde{\mathbf{m}}(\rho,z)_\rho\rangle)]. \tag{D5.24}$$

The oscillating part of the magnetization, $\langle\tilde{\mathbf{m}}(\mathbf{r})\rangle$, can thus be found by first solving Eq. (D5.23) as an ordinary differential equation in ρ to find $\langle\tilde{\mathbf{m}}(\rho,z)_\rho\rangle$ and then evaluating $\langle\tilde{\mathbf{m}}(\rho,z)_z\rangle$ and $\langle\tilde{\mathbf{m}}(\rho,z)_\phi\rangle$ from Eqs. (D5.24) and (D5.20), respectively.

To solve the differential equation (D5.23), we notice that its homogeneous counterpart has solutions ρ^μ with $\mu=-1\pm\nu$. These can be used to construct a Green's function

$$G(\rho,\rho')=(1/2\nu)(\rho')^{-\nu}\rho^{-1+\nu}\quad(\rho<\rho')$$
$$=(1/2\nu)(\rho')^{\nu}\rho^{-1-\nu}\quad(\rho>\rho'), \tag{D5.25}$$

which satisfies

$$\nu^2G(\rho,\rho')-(\partial/\partial\rho)\{\rho(\partial/\partial\rho)[\rho G(\rho,\rho')]\}=\delta(\rho-\rho'), \tag{D5.26}$$

and generates the solution of Eq. (D5.23) as

$$\langle\tilde{\mathbf{m}}(\rho,z)_\rho\rangle=\int d\rho' G(\rho,\rho')(\nu\rho'/c)\langle\tilde{j}(\rho',z)_z\rangle. \tag{D5.27}$$

6. Evaluation for atomic states

a. States of a magnetic ion in free space

For a magnetic ion in any state with standard angular momentum quantum numbers $|L\mathbf{M}_LSM_S)$ or $|LSJM_J)$, the current density has the axially symmetric form indicated in Eq. (D5.15), and the magnetization in the Trammell or MNC gauge can be evaluated by the methods discussed in Appendices D.4 and D.5. This will be illustrated in what follows for one-electron hydrogenic states, for which all results can be evaluated analytically.

The orbital wave function for such a state is

$$\psi_{n,l,m}(\mathbf{r})=R_{n,l}(r)Y_{l,m}(\theta,\phi). \tag{D6.1}$$

The orbital current density is

$$\langle\mathbf{j}_{\mathrm{orb}}(\mathbf{r})\rangle=\mathbf{u}_\phi(2c\mu_A)|R_{n,l}Y_{l,m}|^2m[r\,\sin\theta]^{-1}, \tag{D6.2}$$

where $\mu_{\mathbf{A}}=-\mu_B$ is the anti-Bohr magneton. The spin magnetization in the spin-up state, in the spin-density gauge, is

$$\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_{\rm SD}=\mathbf{u}_z(g_0/2)\mu_A|R_{n,l}(r)Y_{l,m}(\theta,\phi)|^2. \tag{D6.3}$$

The spin-carried electric current density is therefore

$$\langle \mathbf{j}_{\rm spin}(\mathbf{r})\rangle=c\nabla\times\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_{\rm SD}$$
$$=-\mathbf{u}_\phi(g_0/2)\mu_A D_\phi|R_{n,l}(r)Y_{l,m}(\theta,\phi)|^2, \tag{D6.4}$$

where $D_\phi \equiv [\sin\theta(\partial/\partial r)+(1/r)\cos\theta(\partial/\partial\theta)]$. From these forms for $\langle \mathbf{j}_{\rm orb}(\mathbf{r})\rangle$ and $\langle \mathbf{j}_{\rm spin}(\mathbf{r})\rangle$, it follows that $\langle \mathbf{m}_{\rm orb}(\mathbf{r})\rangle$ and $\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle$ are both directed along $\mathbf{u}_\theta \equiv -\mathbf{u}_z\sin\theta+\mathbf{u}_\rho\cos\theta$ in the Trammell gauge and along $\mathbf{u}_z$ in the MNC gauge; $\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_{\rm MNC}$ is of course identical with $\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_{\rm SD}$, which is already perfectly parallel.

To illustrate the relation between the Trammell and MNC gauges, it suffices to consider the 1s state $\psi_{n,l,m}=\psi_{1,0,0}$. The spin magnetization in the MNC gauge is

$$\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_{\rm MNP}=\mathbf{u}_z(g_0/2)(\mu_A/8\pi)e^{-r}. \tag{D6.5}$$

(Here and in all following equations for hydrogenic states, $na_0/2Z$ is taken as the unit of length). The spin-carried electric current density is

$$\langle \mathbf{j}_{\rm spin}(\mathbf{r})\rangle=u_\phi(g_0/2)(\mu_A c/8\pi)\sin\theta e^{-r}, \tag{D6.6}$$

and the spin magnetization in the Trammell gauge is

$$\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_{\rm Tr}=-\mathbf{u}_\theta(g_0/2)(\mu_A/8\pi)[(r+1)/r]\sin\theta e^{-r}. \tag{D6.7}$$

The deviation of this magnetization from perfectly collinear form can be quantified by comparing rms values of its $\parallel$ and $\perp$ parts, which are found to be related by

$$(\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_\perp)_{\rm rms}/(\langle \mathbf{m}_{\rm spin}(\mathbf{r})\rangle_\parallel)_{\rm rms}=1/2. \tag{D6.8}$$

It will be noted that the Trammell magnetization diverges like $1/r$ at small r (although its volume integral is still convergent), which is a consequence of the strong localizing tendency of the Trammell formula. This localizing tendency may be quantified by comparing the values of the mean-square magnetic radius

$$\langle r^2\rangle \equiv \int d^3r r^2|\langle \mathbf{m}(\mathbf{r})\rangle|^2 \Big/ \int d^3r|\langle \mathbf{m}(\mathbf{r})\rangle|^2 \tag{D6.9}$$

yielded by the two gauges, which are found to be related by

$$\langle r^2\rangle_{\rm Tr}/\langle r^2\rangle_{\rm MNC}=7/15. \tag{D6.10}$$

Although the ratios in Eqs. (D6.8) and (D6.10) vary somewhat from state to state, the above values are typical for spin magnetization in other hydrogenic states, as well as for orbital magnetization when it does not vanish. The divergence like $1/r$ is generic for orbital as well as spin magnetization in the Trammell gauge.

For later comparisons we shall also need the MNC orbital magnetization in 3d states. For the state $\psi_{3,2,2}$ one finds

$$\langle \mathbf{j}_{\rm orb}(\mathbf{r})\rangle_{3,2,2}=\mathbf{u}_\phi(\mu_A c/4\pi\cdot 4\cdot 4!)r^3\sin^3\theta e^{-r} \tag{D6.11}$$

and

$$\langle \mathbf{m}_{\rm orb}(\mathbf{r})\rangle_{3,2,2}=\mathbf{u}_z(\mu_A/4\pi\cdot 4\cdot 4!)[r^3(1-\cos^2\theta)$$
$$+r^2(3-\cos^2\theta)+6r+6]e^{-r}. \tag{D6.12}$$

For the state $\psi_{3,2,1}$ one finds

$$\langle \mathbf{j}_{\rm orb}(\mathbf{r})\rangle_{3,2,1}=\mathbf{u}_\phi(\mu_A c/4\pi\cdot 2\cdot 4!)r^3(\sin\theta-\sin^3\theta)e^{-r} \tag{D6.13}$$

and

$$\langle \mathbf{m}_{\rm orb}(\mathbf{r})\rangle_{3,2,1}=\mathbf{u}_z(\mu_A/4\pi\cdot 2\cdot 4!)r^2(r+1)\cos^2\theta e^{-r}. \tag{D6.14}$$

b. States of a magnetic ion in a crystal field

Let us consider a magnetic ion in a crystalline electric field (CEF), which is assumed to have ν-fold rotational symmetry about $\mathbf{u}_z$.

Before turning to model calculations, let us qualitatively examine the effect that such a crystalline electric field can have. Its effect on a single electron, analogous to that of a crystal potential on the linear momentum of a Bloch electron, is to mix values of the azimuthal quantum number by $m\rightarrow m'=m\pm\mu\nu$, where μ is an integer, and where the increment in m is limited by $|m'|\leq l$. In the intermediate or weak CEF scheme (where the crystalline electric field acts on the orbital angular momentum $\mathbf{L}$ of a Hund-coupled n-electron state or on the total angular momentum $\mathbf{J}$), the relevant azimuthal quantum number is M_L or M_J and the limit on its increment is posed by $|M_L|\leq L$ or $|M_J|\leq J$. Such an admixture yields a current density, and hence a magnetization, with terms that depend on the azimuthal angle as $\sin(\mu\nu\phi)$ or $\cos(\mu\nu\phi)$.

For a single d electron in a crystalline electric field with fourfold rotational symmetry, no ϕ dependence arises in first approximation, since the states $|1,m)=|2,\pm2)$ yield nonmagnetic doublets or singlets and other admixtures are forbidden by $|m'|\leq l$. In this approximation the crystalline electric field can only shift the energy of a state without changing its shape, so the orbital magnetization in the MNC gauge remains perfectly collinear. (Small deviations from collinearity can arise from admixtures of other 1 values or from covalent mixing.) In the intermediate CEF scheme, as applied to states with $L=3$ from configurations such as $3d^2$ or $3d^7$, terms proportional to $\sin(4\phi)$ or $\cos(4\phi)$ can arise, but turn out to be rather small. In the weak CEF scheme, as applied to 4f ions, higher harmonics often arise: for example, ions with $J=8$ or 15/2 in a crystalline electric field with fourfold rotation symmetry have current densities including large terms that are sinusoidal in 4ϕ, 8ϕ, and 12ϕ. Since the relevant equations are linear, the contribution of each such harmonic to the magnetization can be calculated by the same methods illustrated below for the first harmonic. The projection methods by which the current density in such n-electron states can be calculated are explained, for example, in Abragam and

Bleaney (1970), but a one-electron example will suffice for the present discussion.

As a concrete example, let us consider a single $3d$ electron in a trigonally oriented cubic crystalline electric field with sign such that the magnetic triplet t_{2g} is the ground level. The relevant orbital eigenstate, whose amplitudes are fixed by symmetry alone (Abragam and Bleaney, 1970), is

$$|\tilde{1},-\tilde{1}\rangle=(2/3)^{1/2}\psi_{3,2,2}-(1/3)^{1/2}\psi_{3,2,-1}. \tag{D6.15}$$

As in Appendix D.5 the current density is divided into parts that are constant and oscillating with respect to ϕ, denoted $\langle \mathbf{j}(\mathbf{r})\rangle$ and $\langle \tilde{\mathbf{j}}(\mathbf{r})\rangle$, respectively. The part $\langle \mathbf{j}(\mathbf{r})\rangle$ arises from expectation values within $\psi_{3,2,2}$ or $\psi_{3,2,-1}$, while the part $\langle \tilde{\mathbf{j}}(\mathbf{r})\rangle$ arises from cross terms. The expectation value for $\psi_{3,2,-1}$ is the negative of that given for $\psi_{3,2,1}$ in Eq. (D6.13). The nonoscillating part of the orbital magnetization is thus a weighted average of the magnetizations evaluated above for angular momentum eigenstates,

$$\langle \overline{\mathbf{m}}_{\mathrm{orb}}(\mathbf{r})\rangle=(2/3)\langle \mathbf{m}_{\mathrm{orb}}(\mathbf{r})\rangle_{3,2,2}-(1/3)\langle \mathbf{m}_{\mathrm{orb}}(\mathbf{r})\rangle_{3,2,1}. \tag{D6.16}$$

Straightforward evaluation shows that the oscillating part of the current density has the form of Eq. (D5.16), with

$$\langle \tilde{j}(\rho,z)_z\rangle=(\mu_A c/4\pi\cdot 12\cdot 4!)2^{1/2}\rho^3 e^{-r}, \tag{D6.17}$$

where $r\equiv(\rho^2+z^2)^{1/2}$, and where the other components are related to this one by Eqs. (D5.21) and (D5.22). The integral for $\langle \tilde{m}(\rho,z)_\rho\rangle$. Eq. (D5.27), can be expressed in terms of elementary functions, but the exact result is lengthy and not very transparent, so for present purposes it seems better to introduce a simplifying approximation. A suitable approximation is obtained by concentrating the whole weight of $\langle \tilde{j}(\rho,z)_z\rangle$ for each z at a single value of ρ,

$$\overline{\rho}\equiv\overline{\rho}(z)\equiv(9+3|z|)^{1/2}, \tag{D6.18}$$

chosen to agree approximately with the maximum of the true $\langle \tilde{j}(\rho,z)_z\rangle$. The approximation is thus

$$\langle \tilde{j}(\rho,z)_z\rangle\approx\langle \tilde{j}(\rho,z)_z\rangle_{\mathrm{approx}}\equiv w(z)\,\delta[\rho-\overline{\rho}(z)], \tag{D6.19}$$

where

$$\begin{aligned} w(z)&\equiv\int_0^\infty d\rho\langle \tilde{j}(\rho,z)_z\rangle\\ &=(\mu_A c/4\pi\cdot 12\cdot 4!)(2)^{1/2}(z^2+3|z|+3)e^{-|z|}. \end{aligned} \tag{D6.20}$$

Inserting this into Eq. (D5.27) yields

$$\begin{aligned} \langle \tilde{m}(\rho,z)_\rho\rangle&=(1/2)[\overline{\rho}(z)]^{-2}\rho^2 w(z)\quad(\rho>\overline{\rho}),\\ &=(1/2)[\overline{\rho}(z)]^{4}\rho^{-4} w(z)\ (\rho<\overline{\rho}). \end{aligned} \tag{D6.21}$$

This simplified approximation solution is accurate for large and small ρ, but it converts the smooth peak of the exact solution for ρ near $\overline{\rho}$ into a sharp peak with discontinuous slope. Equation (D5.20) then yields

$$\langle \tilde{m}(\rho,z)_\phi\rangle=-\langle \tilde{m}(\rho,z)_\rho\rangle. \tag{D6.22}$$

As a result of the crystalline electric field, the magnetization thus indeed develops a noncollinear part even in the MNC gauge. Its relative size can be measured by comparing its rms value to that of the nonoscillating parallel part given by Eq. (D6.16). The result is

$$\{\langle \tilde{\mathbf{m}}(\mathbf{r})\rangle_\perp\}_{\mathrm{rms}}/\{\langle \overline{\mathbf{m}}(\mathbf{r})\rangle_\parallel\}_{\mathrm{rms}}=(2/17)^{1/2}=0.343\ldots, \tag{D6.23}$$

showing that the noncollinear part is indeed quite significant.

APPENDIX E: ANALYSIS OF SCATTERING DATA—LITERATURE

1. Magnetic neutron scattering

The microscopic magnetization in the ferromagnetic $3d$ metals Fe, Co, and Ni was determined from the Bragg scattering of polarized neutrons in the classic experiments of Shull and Yamada (1962), Moon (1964), and Mook (1966). In these papers the data were analyzed by the procedure discussed at the outset of Sec. IV.C.2, which assumes the magnetization to be perfectly parallel.[24]

Moon (1964) emphasized that the assumption that $\langle \mathbf{M}(\mathbf{r})\rangle$ is perfectly collinear, $\langle \mathbf{M}(\mathbf{r})\rangle=\langle \mathbf{M}(\mathbf{r})\rangle_\parallel\equiv\mathbf{u}_z\langle \mathbf{M}(\mathbf{r})_z\rangle$, is a nontrivial one, and Moon and Koehler (1969) made an experimental search for scattering due to $\langle \mathbf{M}(\mathbf{r})\rangle_\perp$ in Co. The experiments were performed with $\mathbf{K}$ along the c axis $\mathbf{u}_\parallel$ of the hcp crystal, since this makes $\langle \mathbf{M}(\mathbf{K})_T\rangle\equiv\langle \mathbf{M}(\mathbf{K})\rangle_\perp$ and means that $\langle \mathbf{M}(\mathbf{K})\rangle_\parallel$ can contribute nothing. Unfortunately, as pointed out by Blume (1970), this choice of $\mathbf{K}$ also means that $\langle \mathbf{M}(\mathbf{K})\rangle_\perp$ must vanish[25] because of the threefold rotation symmetry about $\mathbf{u}_\parallel$, so it is not surprising that the experiment gave a negative result.

As shown in Sec. IV.C.2, the analysis made in the original three articles is exact after all, provided that the MNC gauge is used and the fit is understood to determine only $\langle \mathbf{M}(\mathbf{r})\rangle_\parallel$. Although $\langle \mathbf{M}(\mathbf{r})\rangle_\perp$ cannot be expected to vanish, it contributes nothing to the scattering of neutrons polarized along $\mathbf{u}_\parallel$, not just for $\mathbf{K}$ along $\mathbf{u}_\parallel$ but for any $\mathbf{K}$. Information about $\langle \mathbf{M}(\mathbf{r})\rangle_\perp$ would be of great interest, but its practical determination is an experimental challenge for which no solution is presently in sight.

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[24] In the experimental literature this is often called the Fourier inversion method, and $\langle \mathbf{M}(\mathbf{K})\rangle_\parallel$ or the interpolated $\langle \mathbf{m}(\mathbf{k})\rangle_\parallel$ is usually called the magnetic form factor.

[25] This conclusion is gauge independent because $\langle \mathbf{M}(\mathbf{K})\rangle_\perp$ equals $\langle \mathbf{M}(\mathbf{K})_T\rangle$ and is thus gauge independent, precisely for such $\mathbf{K}$.

Calculations of the scattering of neutrons by n-electron states of an isolated magnetic ion via Racah algebra and related methods have been carried out by Balcar (1975) and Balcar and Lovesey (1989, 1991). Such calculations are less relevant to the central themes of the present article since the value of the scattering cross section, like that of any observable, is independent of the choice of gauge.

A valuable recent review of determinations of the magnetization by magnetic neutron scattering, with further literature references, is that of Brown (1993).

2. Magnetic x-ray scattering

No attempt has been made in this article to review all methods for experimental determination of the microscopic magnetization. One method of great current interest that has not been discussed is the magnetic scattering of x rays, for which the reviews of Balcar and Lovesey (1989), Lovesey (1993), and Lovesey and Collins (1996) should be consulted.

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