

This is the generalization to finite temperatures of the binding energy.^{2,6}

It is a pleasure to thank Dr. P. W. Anderson and Professor J. R. Schrieffer for discussions. We are also grateful to Professor J. Bardeen and Mr. B. D. Josephson for an educational correspondence, and to Professor R. A. Ferrell and Professor R. E. Prange for copies of their work in advance of publication.

*Work supported in part by the U. S. Office of Naval Research and by the Advanced Research Projects Agency.

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MAGNETIC SCATTERING OF NEUTRONS BY NONCOLLINEAR SPIN DENSITIES*

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(Received 1 May 1963)

In the Heitler-London model of a ferromagnet, one associates with each magnetic ion a spin density which points in the direction of the over-all magnetization. This spin density points in the same direction in all parts of the ion. It is entirely possible, however, for a spin density in a ferromagnet to be such that the integral of the density over a unit cell points in the direction of the net magnetization, while the density in different regions of the cell is not collinear with the net magnetization. Such a density might occur in an anisotropic metallic ferromagnet, such as hexagonal cobalt, or in an antiferromagnetic or spiral spin structure. In addition, Overhauser's spin-density-wave¹ theory of metallic magnetism can lead to a density of this form.

It is the purpose of this Letter to point out that it is possible, using neutron scattering techniques, to distinguish noncollinear spin densities from the common variety, and to indicate the simple generalizations of the theory of neutron scattering which are necessary to account for these phenomena. For clarity, we will consider a ferromagnet with one atom per unit cell, but the expressions are easily generalized to more complicated spin structures. We will also consider magnetization densities which arise predominantly from electronic spins, so that orbital scattering can be neglected. To a first approximation the inclusion of orbital effects will not change our results, but taking full

account of orbital moments and anisotropic orbital scattering would complicate the discussion unnecessarily.² Accordingly, we may fix our attention on ferromagnetic cobalt or, with a slight generalization, on antiferromagnetic chromium. The results of the theory may be summarized most easily by pointing out that all of the usual formulas for magnetic scattering³ are still valid on introduction of a noncollinear density, provided that the unit vector which defines the direction of the magnetization is in the new expressions replaced by a unit vector whose direction varies as a function of the scattering vector \vec{K} .

To derive these results we consider first the cross section for elastic magnetic scattering of an unpolarized beam of slow neutrons³:

$$\frac{d\sigma}{d\Omega'} = \left(\frac{\gamma e^2}{mc^2}\right)^2 |\langle q | \sum_i \exp(i\vec{K}\cdot\vec{r}_i) \hat{K} \times (\vec{s}_i \times \hat{K}) | q \rangle|^2. \quad (1)$$

Here $\gamma = -1.91$ is the gyromagnetic ratio of the neutron, and m , \vec{r}_i , and \vec{s}_i are, respectively, the mass, position, and spin of the i th electron in the solid. The summation is over all electrons in the solid, and the matrix element is taken in the ground state $|q\rangle$ of the ferromagnet. $\vec{K} = \vec{k} - \vec{k}'$ is the difference between the initial and final wave vector of the neutron, and \hat{K} is a unit vector in the direction of \vec{K} . Writing out the matrix element

in full, we have

$$\begin{aligned} \langle q | \sum_i \exp(i\vec{K} \cdot \vec{r}_i) \vec{s}_i | q \rangle &= \sum_{\sigma_1 \dots \sigma_\nu} \int_V \dots \int_V d\vec{r}_1 \dots d\vec{r}_\nu \Psi_q^*(\vec{r}_1 \sigma_1, \dots, \vec{r}_\nu \sigma_\nu) \sum_i \exp(i\vec{K} \cdot \vec{r}_i) \vec{s}_i \Psi_q(\vec{r}_1 \sigma_1, \dots, \vec{r}_\nu \sigma_\nu), \\ &= \nu \sum_{\sigma_1 \dots \sigma_\nu} \int_V \dots \int_V \Psi_q^*(\vec{r}_1 \sigma_1, \dots, \vec{r}_\nu \sigma_\nu) \exp(i\vec{K} \cdot \vec{r}_1) \vec{s}_1 \Psi_q(\vec{r}_1 \sigma_1, \dots, \vec{r}_\nu \sigma_\nu) d\vec{r}_1 \dots d\vec{r}_\nu. \end{aligned} \quad (2)$$

Here Ψ_q is the wave function of the solid in the ferromagnetic ground state. The summation is over the values $\pm \frac{1}{2}$ for the spin coordinate of each of the ν electrons in the solid, and the integrations are over the volume V of the entire crystal for each of the ν electrons' position coordinates. In going from the first to the second integral, use

$$\vec{\rho}(\vec{r}_1) = \nu \sum_{\sigma_1 \dots \sigma_\nu} \int_V \dots \int_V d\vec{r}_2 \dots d\vec{r}_\nu \Psi_q^*(\vec{r}_1 \sigma_1, \dots, \vec{r}_\nu \sigma_\nu) \vec{s}_1 \Psi_q(\vec{r}_1 \sigma_1, \dots, \vec{r}_\nu \sigma_\nu). \quad (4)$$

Note that in this expression the integration over \vec{r}_1 is omitted. In the case of a simple ferromagnet, $\vec{\rho}(\vec{r})$ will have the periodicity of the lattice: $\vec{\rho}(\vec{r} + \vec{n}) = \vec{\rho}(\vec{r})$, where \vec{n} is a lattice translation. We may accordingly write (3) as

$$\langle q | \sum_i \exp(i\vec{K} \cdot \vec{r}_i) \vec{s}_i | q \rangle = \sum_{\vec{n}} e^{i\vec{K} \cdot \vec{n}} \int_{V_0} e^{i\vec{K} \cdot \vec{r}} \vec{\rho}(\vec{r}) d\vec{r}, \quad (5)$$

where the integration is now over the volume V_0 of the unit cell. In general, $\vec{\rho}(\vec{r})$ will vary in both magnitude and direction as a function of position in the unit cell. If $\vec{\rho}(\vec{r})$ is collinear, i. e., if it consists of a constant vector times a scalar function of position, we have again the simple situation on the basis of which the standard neutron diffraction formulas have been derived. To see the changes brought about by a more general density, we consider the integral in (5) and write

$$\int_{V_0} e^{i\vec{K} \cdot \vec{r}} \vec{\rho}(\vec{r}) d\vec{r} = S \hat{\eta}(\vec{K}) f(\vec{K}), \quad (6)$$

where $\hat{\eta}(\vec{K})$ is a unit vector in the direction of the integral. The additional condition $f(0) = 1$ then defines S . For $\vec{K} = 0$, the integral becomes

$$\int_{V_0} \vec{\rho}(\vec{r}) d\vec{r} = S \hat{\eta}(0), \quad (7)$$

showing that S is the magnitude of the spin in the unit cell and $\hat{\eta}(0)$ is the direction of the magnetization. We may also interpret $f(\vec{K})$ as the form factor of the spin density in the unit cell. If $\vec{\rho}(\vec{r})$ is

has been made of the antisymmetry of the wave function Ψ_q . We may write (2) more simply as

$$\langle q | \sum_i \exp(i\vec{K} \cdot \vec{r}_i) \vec{s}_i | q \rangle = \int_V \exp(i\vec{K} \cdot \vec{r}_1) \vec{\rho}(\vec{r}_1) d\vec{r}_1, \quad (3)$$

where the spin density $\vec{\rho}(\vec{r}_1)$ is given by the usual quantum mechanical definition,

collinear, then $\hat{\eta}(\vec{K}) = \hat{\eta}(0)$, and the standard results follow. In the general case, $\hat{\eta}(\vec{K})$ need not point in the direction of the magnetization. Physically this is because a neutron scattering experiment looks at the Fourier transform of the spin density. For forward scattering ($\vec{K} = 0$), one observes the total magnetization, i. e., the integral of the spin density over the unit cell. For scattering with nonzero \vec{K} , one weights different parts of the unit cell with the factor $e^{i\vec{K} \cdot \vec{r}}$, so that, if the density points in different directions in different parts of the cell, a magnetization pointing in another direction is observed.

Collecting (6), (5), and (3), we have

$$\langle q | \sum_i \exp(i\vec{K} \cdot \vec{r}_i) \vec{s}_i | q \rangle = \sum_{\vec{n}} e^{i\vec{K} \cdot \vec{n}} S f(\vec{K}) \hat{\eta}(\vec{K}),$$

and

$$\frac{d\sigma}{d\Omega'} = \left(\frac{\gamma e^2}{mc^2} \right)^2 \left| \sum_{\vec{n}} e^{i\vec{K} \cdot \vec{n}} \right|^2 S^2 |f(\vec{K})|^2 \hat{q}^2(\vec{K}), \quad (8)$$

where $\hat{q}(\vec{K}) = \hat{K} \times [\hat{\eta}(\vec{K}) \times \hat{K}]$. This is the same as the usual formula except for the fact that $\hat{\eta}(\vec{K})$ need no longer point in the direction $\hat{\eta}(0)$ of the magnetization. The experimental determination of these phenomena would be accomplished most easily using polarized neutron techniques. The cross section for elastic scattering of a polarized beam by a ferromagnet is

$$\begin{aligned} \frac{d\sigma}{d\Omega'} = & \left| \sum_{\vec{n}} e^{i\vec{K} \cdot \vec{n}} \right|^2 \frac{2}{a^2} + 2a \frac{\gamma e^2}{mc^2} \langle q | \vec{P} \cdot \sum_i \exp(i\vec{K} \cdot \vec{r}_i) \hat{K} \times (\vec{s}_i \times \hat{K}) | q \rangle \sum_{\vec{n}} e^{i\vec{K} \cdot \vec{n}} \\ & + \left(\frac{\gamma e^2}{mc^2} \right)^2 \left| \langle q | \sum_i \exp(i\vec{K} \cdot \vec{r}_i) \hat{K} \times (\vec{s}_i \times \hat{K}) | q \rangle \right|^2, \end{aligned} \quad (9)$$

where a is the nuclear scattering length and \vec{P} is the polarization vector of the neutron beam. On substituting the above expression for the electronic matrix element, we obtain

$$\frac{d\sigma}{d\Omega'} = \left| \sum_{\vec{n}} e^{i\vec{K}\cdot\vec{n}} \right|^2 \left\{ a^2 + 2a \frac{\gamma e^2}{mc^2} S f(\vec{K}) \vec{P} \cdot \vec{q}(\vec{K}) + \left(\frac{\gamma e^2}{mc^2} \right)^2 S^2 |f(\vec{K})|^2 \vec{q}^2(\vec{K}) \right\}. \quad (10)$$

The polarization-dependent term here affords a method of determining the direction of $\vec{q}(\vec{K})$, for the cross section depends on the angle between \vec{P} and $\vec{q}(\vec{K})$. If the cross section is measured for two different reflections as a function of the direction of \vec{P} , the variation in the direction of $\vec{q}(\vec{K})$ due to a noncollinear density can be detected. It is also possible to use the analysis of the direction of polarization of the final beam to detect this effect. The derivation of this is similar to the above and will not be given here.

In addition to being of inherent experimental and theoretical interest, noncollinear spin densities can greatly complicate the interpretation of neutron form-factor measurements and spin-structure determinations. An examination of Eq. (8) shows the sort of difficulties which may arise. Ordinarily one interprets the change in intensity of the scattered beam as a function of \vec{K} to be due to the variation in the form factor. If $\hat{\eta}(\vec{K})$ changes direction, however, as a function of \vec{K} , the factor

$\vec{q}^2(\vec{K})$ will also contribute to the variation in intensity. This will, accordingly, lead to difficulty in accurate determination of form factors, for the separation of these two variations is not possible unless the change of direction of $\vec{q}(\vec{K})$ has been measured in a polarized beam experiment. Similar difficulties occur in spin-structure experiments. Here the absence of certain reflections otherwise expected to occur is taken to indicate $\vec{q}^2 = 0$. According to our discussion, however, the condition $\vec{q}^2 = 0$ determines the direction of $\hat{\eta}(\vec{K})$, which is not necessarily that of the direction of magnetization $\hat{\eta}(0)$. One may be led by this to incorrect conclusions about the direction of magnetization if the presence of noncollinear densities is unsuspected.

I am grateful to H. Alperin, R. Nathans, and S. J. Pickart for pointing out this problem, and for helpful discussions and suggestions.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

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DOUBLE EXCITATION OF HELIUM BY ELECTRONIC IMPACT

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(Received 5 April 1963)

We report a preliminary calculation of the cross section for electron impact excitation of helium to the doubly excited $(2p)^2\ ^3P_g$ state, for which the scattered beam has a maximum of intensity at right angles to the incident beam. Similar calculations for several other states have been performed and will be reported elsewhere. The case considered here is of special interest because it involves the lowest lying, doubly excited state which is stable to autoionization.^{1,2} Thus, while most doubly excited (or "anomalous") states decay by electron ejection within 10^{-13} - 10^{-14} second, helium in the $(2p)^2\ ^3P_g$ state and certain other states can survive for as long as 10^{-9} - 10^{-10} sec-

ond before it ultimately decays by dipole radiation. It is therefore possible for these highly excited atoms to live long enough to participate in chemical reactions analogous to the inverse dissociative-recombination process.

The $(2p)^2\ ^3P_g$ state is the lowest with this symmetry. By representing the angular portions of the wave functions as products of spherical harmonics for the individual electrons, trial functions can be constructed which are orthogonal to all lower states, even the infinite number of $1snp\ ^3P_u$ states. Thus the variation theorem can be applied directly without reference to lower lying states. Therefore energy integrals calculated