where $U_{\mathbf{k},jj'}=\epsilon_j \exp(i\mathbf{k}\cdot\mathbf{r}_j)\delta_{jj'}$. Therefore, the eigenvalues of L_κ(Sáenz) are the same as those of the present $L_\mathbf{k}$, and so the remaining discussion will be carried out in terms of the $L_\mathbf{k}$ and other notation of the present paper.

Sáenz used the result that the eigenvalues of $\epsilon L_\mathbf{k}$ ($\epsilon_{jj'}=\epsilon_j\delta_{jj'}$) are real if $L_\mathbf{k}$ is positive definite; it seems worthwhile to include a proof here. If $L_\mathbf{k}$ is Hermitian and positive definite, then

$L_\mathbf{k}^{-1}$ exists, and is Hermitian and positive definite; (A2)

$L_\mathbf{k}^{1/2}$, $L_\mathbf{k}^{-1/2}$ exist, and are Hermitian and positive definite. (A3)

Now, suppressing the index $\mathbf{k}$, let an eigenvalue of ϵL be μ, with a corresponding eigenvector x:

$$\epsilon L x=\mu x. \tag{A4}$$

Let $Lx=y$; $x=L^{-1}y$:

$$\epsilon y=\mu L^{-1}y. \tag{A5}$$

Take the inner product of each side of (A5) with y to get

$$(y,\epsilon y)=\mu(y,L^{-1}y). \tag{A6}$$

Since ϵ and L^{-1} are both Hermitian, the inner products in (A6) are real, and since L^{-1} is positive definite, $(y,L^{-1}y)\neq 0$. Therefore μ must be real. Hence L positive definite is sufficient (but not necessary) to insure μ real.

Sáenz has also proved, by continuity arguments, that the number of positive (negative) eigenvalues μ is equal to the number of positive (negative) elements in the matrix ϵ, provided $\mu\neq 0$. This can also be proved for each matrix L: $\mathrm{sig}(\epsilon L)=\mathrm{sig}\epsilon$ if L is positive definite (where sig$M\equiv$signature M). Let $L^{1/2}x=z$, $x=L^{-1/2}z$, and (A4) becomes

$$\epsilon L^{1/2}z=\mu L^{-1/2}z. \tag{A7}$$

Multiply on the left by $L^{1/2}$ to get

$$L^{1/2}\epsilon L^{1/2}z=\mu z. \tag{A8}$$

Now $\mathrm{sig}(L^{1/2}\epsilon L^{1/2})=\mathrm{sig}(\epsilon L)$, since both matrices have the same eigenvalue spectrum [(A4) and (A8)]. But $\mathrm{sig}(L^{1/2}\epsilon L^{1/2})=\mathrm{sig}\epsilon$, since the signature is invariant under a conjunctive transformation ($L^{1/2\dagger}=L^{1/2}$). Therefore, $\mathrm{sig}(\epsilon L)=\mathrm{sig}\epsilon$.

It finally remains to show that the eigenvalues found by Sáenz, namely, the absolute values of the (real) eigenvalues μ of ϵL, are the same as those found here. If V is the (unitary) matrix which diagonalizes L, as in Sec. II above, then

$$V^{-1}LV=L_0, \quad V^{-1}L^{1/2}V=L_0^{1/2}, \tag{A9}$$

where L_0, $L_0^{1/2}$ are diagonal matrices whose elements are λ_s, $\lambda_s^{1/2}$, respectively. But the matrix of (A8), whose eigenvalues are μ, is related by a unitary transformation, with V, to the matrix R of Sec. II:

$$V^{-1}L^{1/2}\epsilon L^{1/2}V=L_0^{1/2}V^{-1}\epsilon V L_0^{1/2}=R. \tag{A10}$$

Therefore, the eigenvalues of ϵL are the same as those of R, and this completes the proof. It also follows from (A10) that sig$R=$sigϵ, a result which was used in Sec. III.

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Role of Fermi Surface and Crystal Structure in Theory of Magnetic Metals

DANIEL MATTIS AND WILM E. DONATH
Thomas J. Watson Research Center, International Business Machines Corporation, Yorktown Heights, New York

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We have investigated the magnetic ground state of metals using an idealized theory of magnetism based on the Ruderman-Kittel-Yosida indirect exchange interaction. The preliminary, but suggestive, results reported here are for simple cubic structures and spherical Fermi surface. We find that as the number of electrons is increased, ferromagnetism is replaced by two different antiferromagnetic structures before the Néel state is finally obtained. The first transition occurs at $k_Fa\sim 0.5\pi$ (k_F=Fermi wave vector, a=lattice constant) at which value the rapid changeover occurs, from ferromagnetism to an antiferromagnetic structure of planes of uniform magnetization, which point along alternating directions. These planes lie perpendicular to the (1, 0, 0) axis at first, then abruptly change over to the (1, 1, 0) axis at $k_Fa\sim 0.7\pi$. The new configuration remains the ground state until k_Fa is further increased to 0.85π. But, at that value a final transition occurs to the Néel state. The Néel state (where every spin is surrounded by antiparallel nearest neighbors) then becomes increasingly stable and reaches maximum stability when the Fermi surface touches the zone boundary at $k_Fa=\pi$. It appears from our calculations that the above-mentioned states are the only stable spiral configurations in the simple cubic lattice without the introduction of anisotropy or nonlinearity into the theory.

EVERYONE knows that there are many competing theories of magnetic metals. Each must be judged on its merits in predicting not just the occurrence of ferromagnetism, but also in explaining with a minimum of adjustable parameters the various possible antiferromagnetic orderings.

We have found that an indirect exchange theory (IET) based on the Ruderman-Kittel-Yosida interaction seems satisfactory in this respect when properly considered, and in the present communication we shall present a simple criterion based on this theory. The criterion is that, *all other things being equal*, a three-dimensional metal made of magnetic atoms is ferromagnetic when there are few conduction electrons, or when the magnetic atoms are spatially close, and antiferromagnetic when there are many of them or when the magnetic atoms are far apart. By "antiferromagnetic" we mean to include spiral spin configurations (of which the Néel state is but a special case, when it is allowed by the crystal configuration). In the simplest, isotropic version of the theory applicable to idealized cubic materials, the only parameters which determine the ordering of the magnetic ground state are the product of Fermi wave vector and lattice parameter $k_F a$, and the electronic mean free path l. On the basis of these alone, the theory is capable of distinguishing between the ferromagnetic and various antiferromagnetic orderings in the ground state in a given crystal structure. But the crystalline structure is equally important and we must recognize that, all other things (such as nearest-neighbor spacing) being equal, it is also quite likely for identical magnetic atoms imbedded in different crystallographic arrays to have different magnetic ground states, although there is no simple criterion for this. These conclusions, which are reasonably arrived at on the basis of a model interaction which is long ranged and has nodes, are apparently at variance and invite comparison with any simple analysis on the basis of the nearest-neighbor-overlap description of magnetic interactions.

Before describing the calculation, we must recall briefly some familiar but essential results. In the IET, the magnetic metal is defined as an array of localized spins. These have a direct "exchange" interaction with the delocalized electrons which provide the metallic bonding. The interaction among the localized spins is therefore mainly virtual, by the emission and reabsorption of spin deviations in the conduction medium. The resulting "indirect exchange interaction" is typically long ranged with oscillations characteristic of a sharp Fermi surface; in the simple IET, we assume constant exchange matrix element, spherical Fermi surface, and the rapid convergence of perturbation theory leads one to the Heisenberg-type Hamiltonian for the residual interaction among the localized spins:

$$H=\sum_i \sum_{j\neq i} J(R_{ij})\mathbf{S}_i\cdot\mathbf{S}_j. \tag{1}$$

The effective coupling $J(R_{ij})$ is the famous Ruderman-Kittel-Yosida[1,2] interaction:

$$J(R_{ij})\propto R_{ij}^{-4}[2k_F R_{ij}\cos(2k_F R_{ij})-\sin(2k_F R_{ij})]. \tag{2}$$

[1] M. A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954).
[2] K. Yosida, Phys. Rev. **106**, 893 (1957).

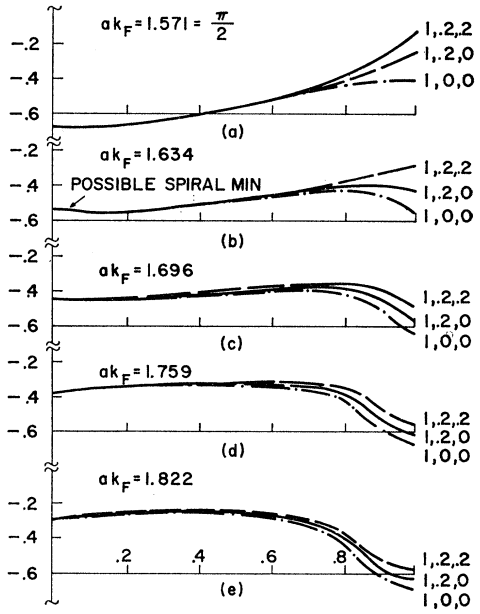

FIG. 1. Finer points of the first ferromagnetic-antiferromagnetic transition [along (100) axis]. Note that (b) indicates the only (and inconclusive) evidence we have found for spiral antiferromagnetism with q_0 not on the principal points of the Brillouin zone boundary or at the origin. Here we plot $F(\mathbf{q})$ along the (1,0,0), (1,0.2,0), and (1,0.2,0.2) direction from the origin to the zone boundary.

However, one should also include an exponential dependence on electron mean free path, which is already well established in the theory of electrical conductivity,[3,4] and multiply this expression by $\exp(-R_{ij}/l)$. In this connection, we should also mention the theory of Bloembergen and Rowland[5] on the indirect coupling mechanism in insulators. They find that (2) must be multiplied by $\exp(-R_{ij}\alpha)$, where α is a factor proportional to the square root of the forbidden energy gap. Although there are at least these two reasons for multiplying Eq. (2) by an exponential decay factor, at present we are reporting on our preliminary results which were found without this factor, and we shall mention the results of subsequent calculations only briefly below.

The Hamiltonian (1) defines quantum—and statistical—mechanical problems which cannot be exactly solved in general. But for the present purposes, a knowledge of the magnetic ground state in the Hartree (product wave function) approximation is sufficient, and this is equivalent to treating the spins as classical vectors. In this approximation the ground state is generally a configuration,

$$S_i^x=S\cos\mathbf{q}_0\cdot\mathbf{R}_i,\quad S_i^y=S\sin\mathbf{q}_0\cdot\mathbf{R}_i,\quad S_i^z=0, \tag{3}$$

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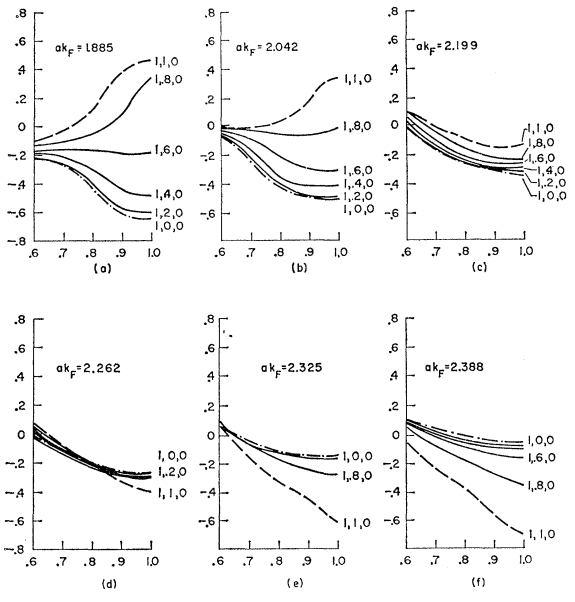

FIG. 2. Finer points of the transition between the spiral $q_0=(\pi,0,0)/a$ structure and the spiral $q_0=(\pi,\pi,0)/a$ structure. It occurs between frames (c) and (d). This is a very rapid transition and may even be discontinuous, as there is no evidence that q_0 assumes intermediate values on the zone boundary. Here $F(\mathbf{q})$ is plotted along various directions, but the position of the curves near the origin (which is now a maximum) is not shown.

leaving $\mathbf{q}_0$ as a vector to be determined below. Interesting limiting cases are ferromagnetism ($\mathbf{q}_0=0$), and the Néel state [$\mathbf{q}_0=a^{-1}(\pi,\pi,\pi)$, in the simple cubic lattice]. The proof that the class of states (3) includes the classical ground state is relatively general, but it is required that the spins form a Bravais lattice.[6] Complications in the hexagonal close-packed structure have, however, been discussed.[7,8] The vector $\mathbf{q}_0$ is the value of $\mathbf{q}$ for which the following lattice sum is a minimum:

$$F(\mathbf{q})=\sum_{\mathbf{R}_i\neq 0} J(R_i)e^{i\mathbf{q}\cdot\mathbf{R}_i}, \tag{4}$$

and it is mainly in this sum that the crystal structure has an effect.[9] Note that values of $F(\mathbf{q})$ away from the minimum also give information, and in principle the energy of elementary excitations may be calculated therefrom. The one case which is exactly soluble is when $q_0=0$, when elementary arguments give for the spin-wave energies,

$$\epsilon_q=2S(F_q-F_0)=Sq^2(d^2F/dq^2)\,|_{q=0}+\cdots, \tag{5}$$

which leads one directly to the Bloch $T^{3/2}$ law, at sufficiently low temperatures.

The function $F(\mathbf{q})$ has topological properties similar to the more familiar energy-band structure and vibration spectra in a lattice. It is defined in the first Brillouin zone, and points of maximum interest are at the center and on the zone boundary. It is therefore quite obvious that the shape of the zone, and therefore, the crystallographic structure, should affect the determination of the ground state as we asserted above. The dependence on crystal structure should be most pronounced when $2k_F$ approaches a reciprocal lattice vector in magnitude.

At the opposite extreme, when $k_Fa\to 0$, the sum (4) can be accurately represented by an integral which shows no dependence on lattice structure at all, and one finds Yosida's result[2] (valid for qa also small)

$$F(q)\sim-\tfrac{1}{2}\left[1+\frac{4k_F^2-q^2}{4k_Fq}\ln\left|\frac{2k_F+q}{2k_F-q}\right|\right], \tag{6}$$

which is, of course, the well-known Fourier transform of $J(R)$. Integrating, instead of summing, always results in ferromagnetism, for which Eq. (5) is applicable. [Common knowledge of this ferromagnetism probably obscured the various antiferromagnetic possibilities of the simple IET. Even among ferromagnetic materials the similarity in spin wave spectra cannot be expected to be as great as predicted by a universal formula (6), for the assumption $k_F\to 0$ is not generally valid.]

Previous work has been reported[8,10,11] which points out various antiferromagnetic possibilities of the interaction (1), but we are reporting on what we believe to be the first systematic investigation of the lattice sum (4), in the simple cubic structure, which we have programmed on the IBM 7090 as a direct lattice sum of up to 6000 points and an approximate integration of the remainder. The results which are displayed are for infinite free path. We have searched the Brillouin zone for the minimum of $F(q)$ for various values of k_F from $0.1\ \pi/a$ to a value π/a where the Fermi sphere touches the zone boundary, and in the figures we present some of the more interesting graphs in self-explanatory form. It is interesting to see the various sorts of antiferromagnetic orderings that occur in the range intermediate between the regions of stable Néel state and stable ferromagnetism.

To summarize the figures, successive minima correspond at first to ferromagnetism then to (100) planes of parallel spins in alternating array, spaced a apart. This is followed (as k_Fa is further increased) by (110) planes of parallel spins in alternating array, the distance between nearest antiparallel planes now being only $a/\sqrt{2}$. This arrangement finally gives way to the Néel configuration, the "most antiferromagnetic" of all. There are no minima except at points of high symmetry, and the transitions are rather abrupt.

These results are completely confirmed by calcula-

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[7] T. A. Kaplan, Phys. Rev. **124**, 329 (1961).
[8] E. J. Woll and S. J. Nettel, Phys. Rev. **123**, 796 (1961).
[9] In cubic structures with one spin per magnetic cell, this sum is real. Also, q can be reduced to the first Brillouin zone. The classical theory for the hexagonal structure is given in reference 7, and the spin-wave theory in reference 8.
[10] P. G. DeGennes, Compt. rend. **247**, 1836 (1958).
[11] J. Chevalier and W. Baltensperger, Helv. Phys. Acta **34**, 859 (1961).

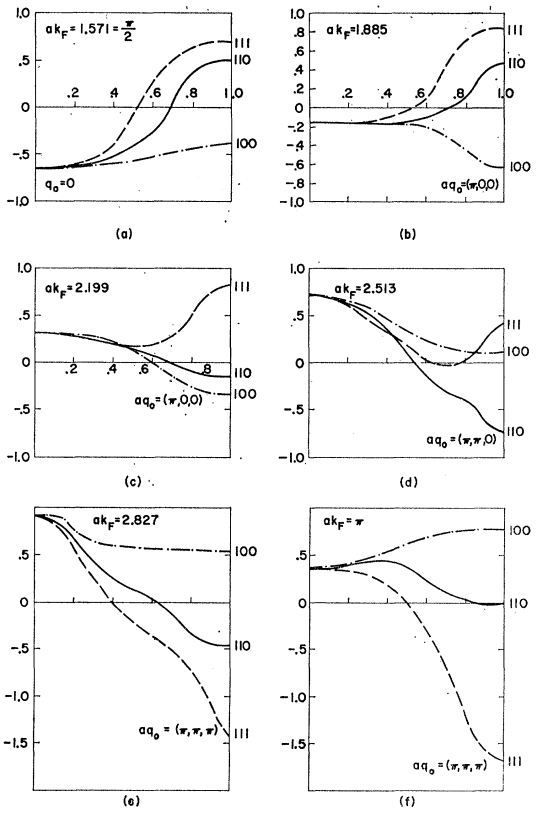

FIG. 3. Plot of

$$F(\mathbf{q}) \equiv a^3 \sum_{i \neq 0} [\exp(i\mathbf{q}\cdot\mathbf{R}_i)](2k_F R_i \cos 2k_F R_i - \sin 2k_F R_i)(8\pi k_F R_i^4)^{-1},$$

along the three principal directions for the simple cubic structure, from the origin to the zone boundary. Note how the minimum at q_0, which determines the ground-state spin configuration, moves with increasing $k_F a$. The ferromagnetic state ($q_0=0$) is stable in frame (a) and for all values of $k_F a$ smaller then $\pi/2$. The Néel state first occurs in frame (e) and is, of course, much stabler at (f).

tions using a finite mean free path, which converge faster and are therefore the most reliable. On the basis of such calculations, we conclude that the dubious minimum in Fig. 1(b) can definitely be disregarded, and we can consider it established that in the simple cubic structure the planes of parallel spins coincide with principal crystallographic planes, within the accuracy and the validity of Eq. (1). We plan to present the results of the finite free path calculations completely at a future date, in conjunction with other interesting features of this investigation which are not germane to the present discussion. For the present, suffice it to say that although a finite free path does slightly affect the ranges of $k_F a$ over which the various configurations are stable, it does not change the abruptness with which the transitions occur, e.g., as shown in Fig. 2.

A surprising result of the calculation is that for $k_F a = \pi/2$ only, all contributions to the lattice sum except from nearest neighbors apparently canceled out to good accuracy, and this feature persisted at all values of the free path. Thus, in comparing Fig. 3(a) with a nearest-neighbor spectrum

$$F(q_x,q_y,q_z) = -\text{const}(\cos q_x a + \cos q_y a + \cos q_z a), \quad (7)$$

we find this formula fits the curve to within an accuracy of 10%. However, this does not necessarily mean that a magnetic substance with $k_F a = \pi/2$ is indistinguishable from one with nearest-neighbor interactions, although they can reasonably be expected to share many similar properties at low temperatures where spin-wave theory is applicable.

The interaction of Eq. (2) which we have investigated has qualitative merit in that it has the oscillatory behavior expected in metals,[12] and depends on a parameter which presumably can be obtained experimentally by other means. However, it does not serve as a quantitative theory of magnetic behavior, for it does indeed depend on a series of approximations which are not universally valid. We also have neglected effects of, e.g., electromagnetic, and anisotropic interactions,[7,13,14] all of which must be pieced together in order to understand the magnetic properties of a specific substance. However, we hope to have made apparent the many possibilities of the simple IET in magnetic metals which future refinements should amplify and clarify.

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