GENERALIZED SUSCEPTIBILITIES AND MAGNETIC ORDERING OF HEAVY RARE EARTHS*

W. E. Evenson and S. H. Liu
Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa 50010
(Received 11 April 1968)

The generalized magnetic susceptibilities, $\chi(\vec{q})$, have been calculated along the line Γ to A for the heavy rare-earth metals, Gd, Dy, Er, and Lu, using energy bands previously calculated by Keeton and Loucks. The initial ordering periodicity for each of these metals can be related to Fermi-surface geometry by examining the maxima in our susceptibilities. Agreement with experimental magnetic wave vectors is satisfactory.

The Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange interaction can easily be related to a $\vec{q}$-dependent susceptibility, $\chi(\vec{q})$. This generalized susceptibility is the same as is obtained from the Kubo[1] linear-response formalism, and for one atom per unit cell it has the form[2]

$$\chi(\vec{q}) = \frac{1}{N}\sum_{\vec{k},n,n'} \frac{f_{\vec{k}n}(1-f_{\vec{k}+\vec{q}+\vec{K}_0 n'})}{E_{n'}(\vec{k}+\vec{q}+\vec{K}_0)-E_n(\vec{k})}, \qquad (1)$$

where the $f_{\vec{k}n}$ are Fermi-Dirac distribution functions for reduced wave vector $\vec{k}$ and band n, the $E_n(\vec{k})$ are the energy bands, $\vec{K}_0$ is the reciprocal lattice vector necessary to reduce $\vec{k}+\vec{q}$, and we have assumed matrix elements to be constant and have factored them out.

In previous work the evaluation of $\chi(\vec{q})$ has been done using free-electron energy bands. The band calculations on rare-earth metals[3] show that they are not free-electron-like, but resemble more closely the transition element bands. Also, the Fermi surfaces are very nonspherical. We feel, therefore, that the use of free-electron bands has been the weakest point in previous calculations. In this work we have kept all the usual approximations[2] in the derivation of Eq. (1), but we have put in realistic bands. Our results show that the Fermi surface geometry is the dominant factor in the determination of the magnetic ordering periodicity in the heavy rare earths.

We are interested only in $\vec{q}$ along the line Γ to A of the Brillouin zone because all the magnetic ordering structures observed in the heavy rare earths can be described by a wave vector in that direction.[4] The metals we are interested in all have hcp crystal structures, and for $\vec{q}$ in the special direction Γ to A, we can treat $\chi(\vec{q})$ in the double-zone representation. (Hereafter, q will indicate the magnitude of the vector $\vec{q}$ with $\vec{q}$ restricted to the line Γ to A.) Relativistic-augmented-plane-wave energy bands for Gd, Dy, Er, and Lu were available to us from the calculations performed by Keeton and Loucks.[3] We have used these bands to calculate the susceptibilities of these four metals. (These are paramagnetic bands, so our conclusions apply to the initial ordering of the metals, before the bands are too greatly perturbed by the magnetic interactions.) We have used these bands in the double-zone representation, where Eq. (1) is the correct formulation of the susceptibility, because the maximum splittings introduced on the AHL zone face were within the numerical accuracy of the bands. The relativistic form of the bands was important to the present results, however, because of significant changes in relative positions of the bands and in the Fermi surface introduced by the relativistic formulation.[3,5]

The maximum in susceptibility determines the stable magnetic structure.[2] It was originally pointed out by Lomer[6] that if the Fermi surfaces may be approximated by pieces of parallel planes separated by a wave vector Q, then there occurs a logarithmic divergence in $\chi(\vec{q})$ at $q=Q$. This is generally referred to as "nesting Fermi surfaces." The relation between the shape of $\chi(\vec{q})$ and the Fermi surface geometry was discussed in detail by Roth, Zeiger, and Kaplan.[7] One may conclude from their discussion that the necessary condition for a maximum in $\chi(\vec{q})$ is to have sizable areas of Fermi surface that can nest with roughly the same wave vector.

To perform the numerical calculation of Eq. (1), we have neglected the temperature dependence of the Fermi functions, and we have used the three-dimensional extension of the trapezoidal rule to perform the integration over $\vec{k}$. We have used a mesh containing 27 216 points in the Brillouin zone for this integration. The finite mesh may introduce spurious peaks in the susceptibility, and is the principal source of noise in our calculation. Comparison with the Fermi surface geometry allows us to eliminate peaks due only to the numerical procedures since the

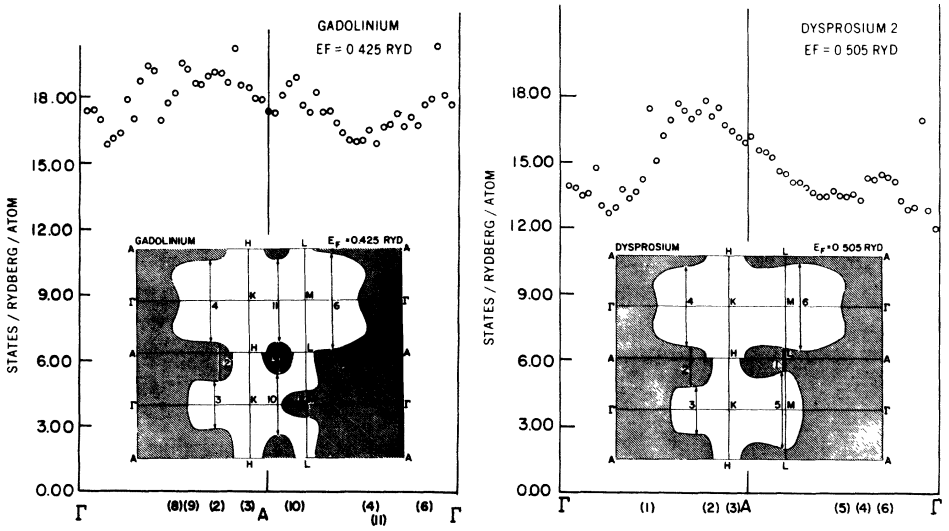

FIG. 1. Generalized susceptibilities and Fermi-surface cross sections for gadolinium and dysprosium.

Fermi surface is what is critical in determining the shape of $\chi(\vec{q})$.

In considering the convergence of the sums on energy bands indicated in Eq. (1), we calculated the susceptibility for Dy (using the bands for the potential called Dy 2 in Ref. 3) using (1) the eight calculated bands, (2) the eight calculated bands plus four free-electron bands above them, and (3) only the two bands which determine the Fermi surface. We found no appreciable difference in the features of the susceptibility in these three cases, only an essentially q-independent shift. The reason for this seems to be that the bands near the Fermi energy are quite flat. For those bands which are much higher or lower than the Fermi energy, the energy denominators of Eq. (1) become large and the contributions to the susceptibility become less important. In view of this result, the calculations that are reported here refer only to the two bands determining the Fermi surface.

We have considered also the effect of varying the Fermi energy up or down by 0.005 Ry. We find that the Fermi surfaces change very gradually along the heavy rare-earth series when we do this. For example, the Fermi surfaces for Gd with E_F decreased by 0.005 Ry and for Dy are almost identical. There are subtle differences, however, that would prevent us from simply taking one set of bands and varying the Fermi energy to get all the susceptibilities of the heavy rare earths. In general, increasing the Fermi energy

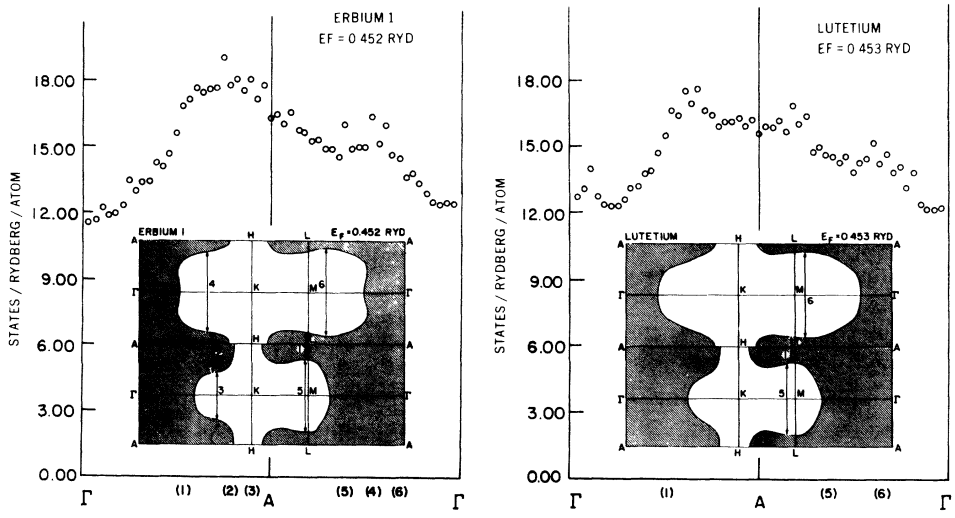

FIG. 2. Generalized susceptibilities and Fermi-surface cross sections for erbium and lutetium.

for a given set of bands decreases the wave vector Q at which the maximum in the susceptibility occurs.

In Figs. 1 and 2 we show the calculated susceptibilities along with appropriate Fermi-surface cross sections for Gd, Dy, Er, and Lu. (The potentials called Dy 2 and Er 1 in Ref. 3 are those used here for Dy and Er.) When only the bands defining the Fermi surface are used, it is straightforward to show that $\chi(0) = \frac{1}{2}N(E_F)$, where $N(E_F)$ is the density of states at the Fermi energy. As an internal check, we note that the limit of $\chi(\vec{q})$ as q goes to zero is in good agreement with $\frac{1}{2}N(E_F)$ from Keeton and Loucks[3] in each case. We see that the susceptibilities for Dy, Er, and Lu have well-defined peaks around $0.6\pi/c$. The magnetic wave vector for these metals is slightly smaller than this, being (in units of π/c) 0.49 for Dy, 0.57 for Er, and 0.53 for Lu by extrapolation from Tb-Lu alloys.[4] Small increases in Fermi energy without changing the bands can bring the positions of the maxima in the susceptibilities into agreement with the experimental values. However, there is at present no reason to expect any better agreement than we have obtained using the calculated Fermi energies. The inclusion of a q-dependent matrix element could have the effect of pulling down the right side of the susceptibilities shown in Figs. 1 and 2 and hence shift the peak slightly to the left into better agreement with experiment. Also, the energy bands are not sufficiently well known at present to expect detailed quantitative agreement with experiment.

We note that the beginning of the peaks in the susceptibilities of Dy, Er, and Lu corresponds quite well to the Fermi-surface separation we have labeled (1). This piece between two arms of the Fermi surface has been called the "webbing" by Keeton and Loucks.[3] They have suggested that the webbing may be responsible for the antiferromagnetic ordering arrangements in the heavy rare earths. Our results seem to confirm this idea. The webbing seems to be important to determining the exact position of the peak in Dy, Er, and Lu. If we look at Gd where the webbing is absent, we see that there is a much wider range of q's that contribute significantly to the susceptibility, so that $\chi(\vec{q})$ is rather flat. A q-dependent matrix element would probably pull down the right of the curve and establish $q = 0$ quite firmly as the maximum of χ, in agreement with the ferromagnetism observed in Gd at its initial ordering point. The webbing in the heavier metals tends to allow a cluster of q's in a small range around the webbing q to dominate the susceptibility.

The good agreement between the experimental and the calculated magnetic ordering periodicity indicates that most of the magnetic ordering information is contained in the energy bands, and the matrix elements are probably smooth functions of q. When we allow the matrix elements to be smoothly decreasing functions of q, we see that our curves are in nice qualitative agreement with the experimental magnon spectrum obtained for Tb-10% Ho by Møller, Houmann, and Mackintosh.[8] We get a very nice peak in each half of the susceptibility curves just as their analysis showed

More experimental and theoretical work will have to be done before the susceptibilities can be understood in greater detail. The inherent uncertainties in calculations such as ours are presently rather large because of the uncertainties in the band calculations and of our lack of understanding of the matrix elements that enter the indirect-exchange interaction.

We wish to acknowledge the kindness of Dr. S. C. Keeton and Dr. T. L. Loucks in providing the energy bands used in this calculation. We have also benefited greatly from many discussions with Dr. Loucks during the course of this work.

---

*Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.

†Danforth Graduate Fellow.

[1]R. Kubo, J. Phys. Soc. Japan 12, 570 (1957).

[2]A good reference for the formal development of the theory is T. Kasuya in Magnetism, edited by G. T. Rado and H. Suhl (Academic Press, Inc., New York, 1966), Vol. II(B), pp. 215-294.

[3]S. C. Keeton and T. L. Loucks, Phys. Rev. 168, 672 (1968), and references cited therein.

[4]W. C. Koehler, J. Appl. Phys. 36, 1078 (1965).

[5]S. C. Keeton, thesis, Iowa State University, 1966 (unpublished).

[6]W. M. Lomer, Proc. Phys. Soc. (London) 80, 489 (1962).

[7]L. M. Roth, H. J. Zeiger, and T. A. Kaplan, Phys. Rev. 149, 519 (1966).

[8]H. Bjerrum Møller, J. C. G. Houmann, and A. R. Mackintosh, Phys. Rev. Letters 19, 312 (1967).

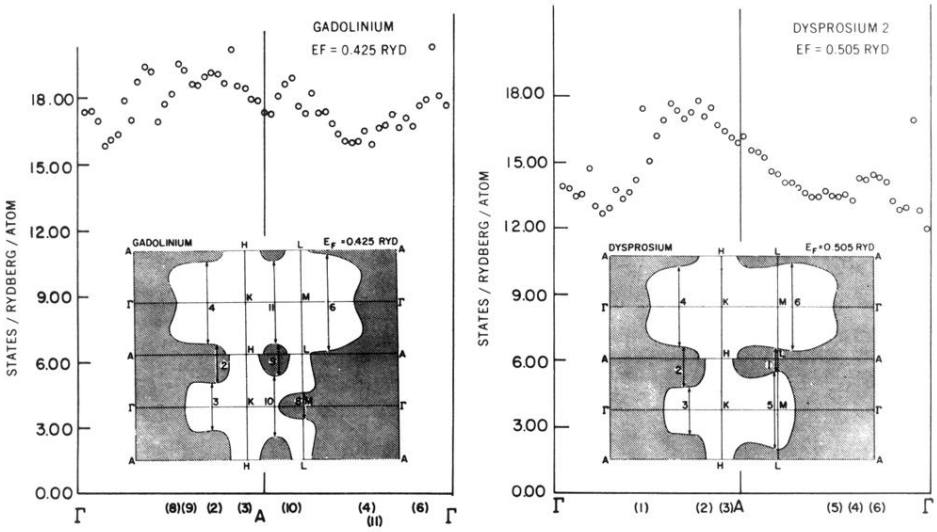

FIG. 1. Generalized susceptibilities and Fermi-surface cross sections for gadolinium and dysprosium.

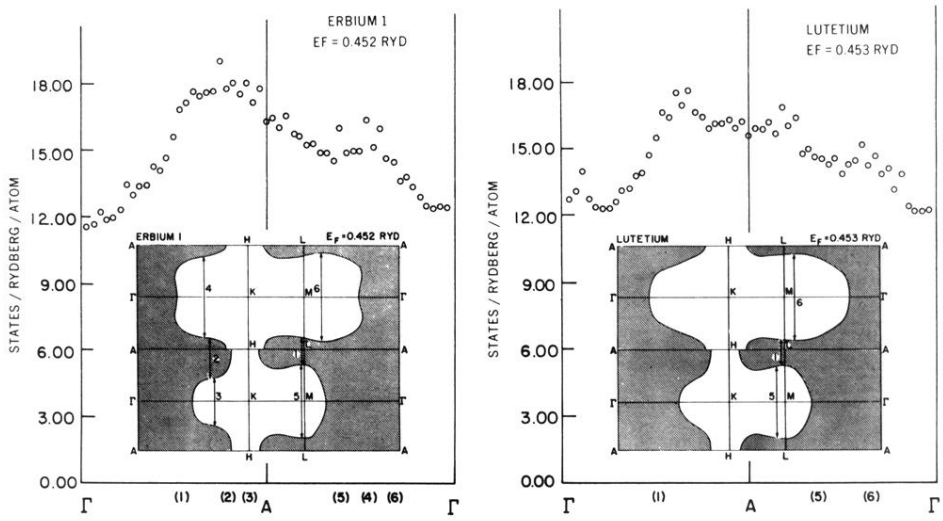

FIG. 2. Generalized susceptibilities and Fermi-surface cross sections for erbium and lutetium.