appreciates the considerable distance still separating theory and experiment even for these relatively simple cases. We suggest that the Au(Yb) system, which displays clear covalent-mixing character and has a relatively simple electronic structure, and for which there is now an extensive body of experimental data, is an excellent candidate for further theoretical work.

We thank Dr. P. T. Coleridge for providing the orbital weighting factors for the Au Fermi surface, Dr. Yun Chung for considerable help with crystal growth, and Professor R. J. Higgins for several helpful discussions. This research was carried out in part with support from the National Science Foundation through Grants No. DMR 74-07652 A02 and No. DMR 78-06271, and was performed in part under the auspices of the U. S. Department of Energy.

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Planar Coupling Mechanism Explaining Anomalous Magnetic Structures in Cerium and Actinide Intermetallics

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(Received 4 December 1979)

Cerium and light actinide monopnictides of NaCl structure have extraordinarily strong magnetic anisotropy, with unusual magnetic structures and transitions. We show that this behavior can be understood on the basis of a Coqblin-Schrieffer–type interaction, effectively treating mixing of f and conduction electrons.

PACS numbers: 75.10.Lp, 75.25.+z, 75.30.Et, 75.30.Gw

Cerium and light actinide monopnictides of NaCl structure show remarkably anisotropic magnetic properties[1] favoring $\langle 100 \rangle$ alignment, and exhibiting transitions between unusual linear magnetic structures. An important feature[2] is the extreme anisotropy of the moment correlations in USb, showing stronger interactions within ferromagnetic sheets than between them.

The peculiar magnetic structural behavior and transitions cannot be explained by the previously considered[3] crystal-field distortional effects, but requires[1] an anisotropic magnetic coupling of a special sort. This led us to consider[1] the presence of a Coqblin-Schrieffer–type (CS) interaction,[4] taking into account combined spin and orbital exchange scattering of the conduction electrons by the localized f moments. Such an interaction effectively treats the covalent mixing of f and conduction electrons.[5] It is physically reasonable to expect such hybridization effects involving $4f$ or $5f$ electrons to be much more important in cerium and light actinide intermetallics than for heavier rare earths or actinides.

We have found a significant correction[6] to the form found by Coqblin and Schrieffer for the cerium-cerium interaction. (This involves no change in the treatment of Coqblin and Schrieffer for the conduction-electron interaction with the localized f electrons.) This corrected form for the CS cerium-cerium interaction makes clear the basis for the anomalous magnetic behavior of the cerium and light actinide monopnictides.

Coqblin and Schrieffer[4] show that the interaction between a pair of Ce^{3+} ions separated by R_{ij} is

$$V_{ij}=\sum_{M,M'} E^{M,M'}(R_{ij})\Big(c_{M'}{}^{\dagger}(i)c_M(i)-\tfrac{1}{6}\delta_{MM'}\sum_{M''} n_{M''}(i)\Big)\Big(c_M{}^{\dagger}(j)c_{M'}(j)-\tfrac{1}{6}\delta_{MM'}\sum_{M''} n_{M''}(j)\Big), \tag{1}$$

Here $c_{M'}{}^{\dagger}(i)$ and $c_M(i)$ create and destroy states $|M'\rangle$ and $|M\rangle$, respectively, at site i; and $n_{M''}(i)$ is the number operator at site i. The axis of quantization is the bonding axis joining the coupled sites. The sums in (1) are over the six $J=\frac{5}{2}$ free-ion states. The coupling constant $E^{MM'}(R_{ij})$ is given by[4]

$$E^{MM'}(R_{ij})=2\sum_{k\sigma k'\sigma'}\frac{f_k(1-f_{k'})}{\epsilon_k-\epsilon_{k'}}\,|\mathcal{J}^{MM'}_{k\sigma k'\sigma'}|^2\cos[(\vec{k}-\vec{k}')\cdot\vec{R}_{ij}]. \tag{2}$$

The difference between the CS interaction and the conventional Ruderman-Kittel interaction occurs because of the dependence of the exchange parameter, $\mathcal{J}^{MM'}_{k\sigma k'\sigma'}$, on M and M' as given by Eq. (17) of Ref. 4.

The error in Ref. 4 occurred in the course of expanding the plane waves in terms of spherical partial waves. However, such an expansion is unnecessary. In fact, the angular integration [i.e., integrals over polar angles θ_k and $\theta_{k'}$ in Eq. (2)] can be expressed exactly in terms of sums of products of integrals of the form

$$\mathcal{g}_{|M\pm 1/2|}\equiv\int_{-1}^{1}dx\,e^{ikRx}(P_3{}^{|M\pm 1/2|})^2, \tag{3}$$

where $P_3{}^{|M\pm 1/2|}$ is an associated Legendre polynomial. To lowest order in $1/kR$ [giving the lowest order in $1/k_FR$ for $E^{MM'}(R)$], the only nonvanishing integrals are for $M=\pm\frac{1}{2}$. *Physically this says that the coupling, to lowest order in* $1/k_FR$, *comes from the* f *orbital states with* $m=0$, *i.e., those that pile up charge along the bonding axis.*

Thus,

$$E^{MM'}(R_{ij})=F(R_{ij})G(M,M'), \tag{4}$$

where the only nonvanishing $G(M,M')$ are for M and M' equal to $\pm\frac{1}{2}$; and $G(M,M')$ has the same value for each of the four possible combinations of $M,M'=\pm\frac{1}{2}$. [The range function $F(R_{ij})$ is the same as that for the conventional Ruderman-Kittel interaction.] This gives a cerium-cerium coupling having strong anisotropy with respect to the bonding axis.

The operators in Eq. (1) are defined with respect to a coordinate system with quantization (z) axis directed along pair bonding axis $\vec{R}_{ij}$. To consider magnetic ordering, it is convenient to express each pair interaction with respect to a coordinate system with the direction of ordered moment as quantization axis. After this transformation, the interaction Hamiltonian of a cerium ion (labeled $j=0$) with the surrounding lattice of Ce ions (approximated by thermal averages of operators) is given by

$$\mathcal{H}=-\textstyle\sum_i E(R_{i0})\sum_\epsilon\sum_\sigma\sum_\mu\sum_\nu B^{\epsilon\sigma}_{\mu\nu}(i)\langle c_\epsilon{}^{\dagger}(i)c_\sigma(i)\rangle c_\mu{}^{\dagger}(0)c_\nu(0). \tag{5}$$

The upper indices on B label the states of a neighbor, and the lower indices label the states of the central site.

Here we shall point out the principal physical characteristics of the transformed interaction (5), and how it explains striking features in the experimental behavior. This is most easily done by referring to Fig. 1 giving the diagonal elements $B^{5/2,5/2}_{5/2,5/2}(\theta)$ and $B^{-5/2,-5/2}_{5/2,5/2}(\theta)$, when the moments are saturated for parallel and antiparallel alignment, respectively, of neighbor and central site moment. θ is the angle between the bonding axis, connecting site i to the central site and the magnetic axis. (For a Heisenberg interaction, these matrix elements would be constant for varying θ, with one the negative of the other.)

There are two features of interest in Fig. 1. First is the strong anisotropy, giving weakest coupling along the magnetic axis and strongest coupling in the transverse plane. (This occurs because the CS coupling comes from orbital states that pile up charge along the bonding axis, while orbital moment is maximized for states where the charge is piled up in the plane transverse to the moment.) This explains the observed anisotropy in coupling,[2] giving essentially planar ordering.

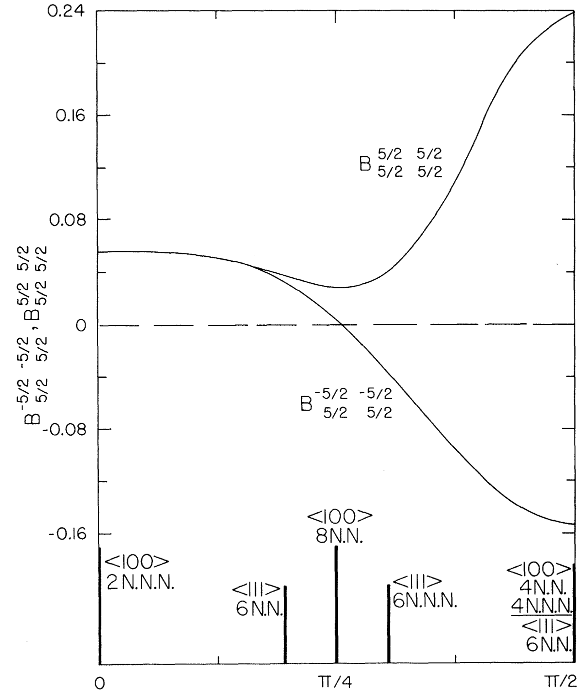

FIG. 1. Variation of diagonal matrix elements for coupling between Ce^{3+} ions with saturated moment, vs angle (θ) between bonding axis and axis of magnetic ordering. The bars at the bottom of the figure identify the values of θ for the nearest (N.N.) and next-nearest (N.N.N.) neighbors for moment along ⟨100⟩ and ⟨111⟩, e.g., "4 N.N." under "⟨100⟩" next to the bar at $\theta=\frac{1}{2}\pi$ means that for ⟨100⟩ moment alignment, four nearest-neighbor bonding axes are at $\theta=\frac{1}{2}\pi$ with respect to the moment direction.

The second feature of interest in Fig. 1 is that $B^{-5/2,-5/2}_{5/2,5/2}(\theta) \neq -B^{5/2,5/2}_{5/2,5/2}(\theta)$. This gives an inherent advantage to ferromagnetic ordering. This asymmetry favoring ferromagnetism follows from the fact that the magnetic quantum numbers of the conduction and localized electrons are exchanged in the scattering involved in the CS mechanism. This exchange of magnetic quantum numbers is preserved on performing the second-order perturbation theory giving the Ce-Ce interaction. This exchange of magnetic quantum numbers means that a term diagonal in M (for quantum axis along bonding axis) on one site couples preferentially to a term diagonal in the same M on the other site.

The fact that the ordering within a $\{100\}$ plane is ferromagnetic for all cerium and light actinide monopnictides studied, indicates that $E(R_{i0})$, at least on average, is ferromagnetic (or sufficiently close to ferromagnetic so that the inherent matrix-element effect favoring ferromagnetism tips the balance).

Once one recognizes $E(R_{i0})$ as being positive, the physical basis for the unusual magnetic structural behavior of CeBi, CdSb, and other monopnictides of interest, becomes clear. Basically

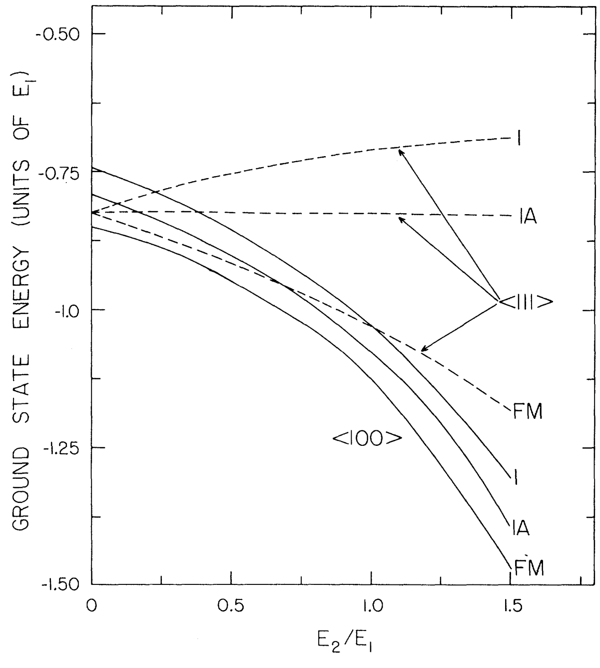

FIG. 2. Ground-state energy vs ratio of next-nearest to nearest-neighbor CS coupling for different magnetic structures with ⟨100⟩ or ⟨111⟩ alignment. (FM denotes ferromagnetism.)

one wants as many neighbors as possible aligned parallel to a given Ce ion and in the angularly favored bonding axis orientation (relative to the magnetic axis) region (near $\theta=\frac{1}{2}\pi$ in Fig. 1).

As shown in Fig. 2, this gives a preference for $\langle 100\rangle$, compared to $\langle 111\rangle$, alignment for a wide range of ratio of next-nearest to nearest-neighbor coupling. (Why this is so can easily be seen from Fig. 1 for the case of nearest and next-nearest interactions approximately equal. $\langle 100\rangle$ alignment gives more neighbors in the angularly favorable region. Variation of the ordered moment at $T=0$ as E_2/E_1 changes also enters into the behavior of Fig. 2.) Figure 2 also shows the relative preference for various types of magnetic ordering. The type-I*A* and -I antiferromagnetic orderings are pertinent to CeBi. They are an up-up-down-down and an up-down-up-down sequence of planar moments, respectively (i.e., moments within individual planes transverse to the magnetic axis are ferromagnetically aligned). Experimentally, CeBi has a first-order transition close to $\frac{1}{2}T_{\rm N}$, from type-I*A* to type-I antiferromagnetic ordering with increasing temperature. This occurs close to magnetic saturation with at most a very small change in ordered moment and size of tetragonal distortion.

As shown in Fig. 2, ferromagnetic ordering along $\langle 100\rangle$ is slightly favored over type-I*A* antiferromagnetism, which in turn is slightly favored over type-I antiferromagnetism. The reason for this is that a plane having moments point "up" the magnetic axis because of the angular variation of coupling shown in Fig. 1 favors having as many planes as possible that are also "up" as close to it as possible. Quite important is that the difference in energy between the ferromagnetic, I*A*, and I states is quite small compared to their difference from the disordered state (zero of energy in Fig. 2.)

One expects a small Heisenberg interaction to be present[4] in addition to the CS interaction. If this is antiferromagnetic, then the small energy advantage of the ferromagnetic ordering can disappear, leaving the I*A* and I antiferromagnetic states to compete. Detailed calculations[7] along this line give model behavior for CeBi, shown in Fig. 3, where the I*A*-to-I transition occurs close to saturation with extremely small changes in distortion and magnetic moment, allowing agreement with experiment. Such behavior has proved impossible to obtain with other types of interaction.[1]

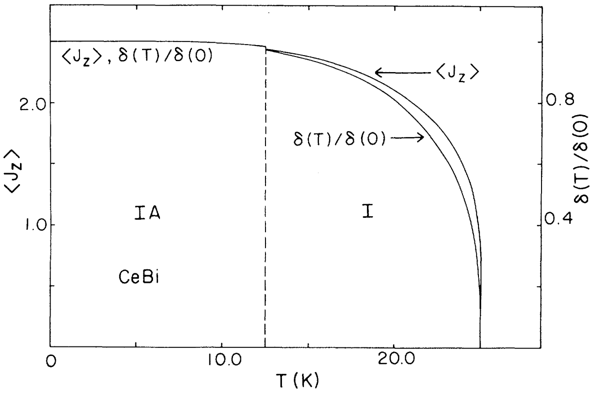

FIG. 3. Expectation value of J_z (7/6 of ordered moment per $\rm Ce^{3+}$) and relative distortion, vs temperature, from model calculation for CeBi.

The CS interaction also makes it favorable to develop disordering through the appearance of nonmagnetic planes as occurs[8] in CeSb. A nonmagnetic plane acts like half an "up" plane plus half a "down" plane. Then for the CS interaction, an up plane can prefer having a nonmagnetic and a down plane, as opposed to two down planes, as neighbors. Detailed theory shows that the tendency to have nonmagnetic planes increases as the crystal-field splitting increases relative to $kT_{\rm N}$. Thus it is reasonable for nonmagnetic planes to occur in CeSb and not in CeBi. The CS interaction also gives an apparent temperature dependence to the crystal-field splitting above $T_{\rm N}$ as observed experimentally.[9]

We appreciate interesting discussion with G. H. Lander and S. K. Sinha. We are grateful to B. Coqblin and J. R. Schrieffer for confirming our correction to the CS interaction. This work was supported in part by the National Science Foundation, Grant No. DMR-79-07069.

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Observation of the Approach to a Polarization Catastrophe

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(Received 24 January 1980)

Optical measurements of phosphorus-doped silicon yield a donor susceptibility which can be fitted with a critical form that extrapolates to a polarization catastrophe at the insulator-metal transition. The exponent is about twice classical predictions and demonstrates the quantum nature of the transition.

PACS numbers: 71.30.+h

As an ensemble of atoms approach one another, a transition from an insulating to a metallic state can occur.[1] A classic example in which such a transition occurs is the random array of impurity atoms in a crystalline semiconductor. Analysis of a variety of measurements on such materials[1] has indicated that at low impurity concentrations the electrons are localized. The behavior of the dc electrical conductivity is nonanalytic at zero temperature, being zero over a finite range (in density) in the insulator and finite in the metal, but the conductivity varies smoothly at finite temperatures. The behavior of the donor electric susceptibility χ is less clear. (χ is related to the total dielectric constant, ϵ, and that of the host Si, ϵ_{Si}, through $\epsilon = \epsilon_{\text{Si}} + 4\pi\chi$.) In the insulator, χ should be finite and positive,[2] and in the metal it may be either large and negative, as indicated by the Drude theory, or infinite, as suggested by Altshuler and Aronov[3] for a disordered system.

Classical analyses[4,5] based on percolation theory for metallic inclusions in a dielectric predict a critical divergence of χ, with various values of the exponent. In contrast to this prediction of a polarization catastrophe, Ghazali and Leroux-Hugon[6] find that χ is enhanced by only a factor of 3 (relative to the result for noninteracting donors) for their model of a doped semiconductor in which the donors are arranged on a lattice.

The experimental behavior of χ is also the subject of some disagreement. In 1956, D'Altroy and Fan[7] found a large enhancement in χ at microwave frequencies and low temperature for doped Ge. Measurements of metal ammonia solutions[8] near room temperature suggested only a small enhancement and a value of $\chi = 0$ at the critical density, n_{MI}. Reflectivity of doped Ge at two far-infrared frequencies and low temperatures was interpreted[9] as indicating no enhancement whatever of χ in the insulating phase. In contrast, Castner *et al.*[2] found enhancements up to a factor of 3 based on low-frequency capacitance measurements extrapolated to zero temperature for several dopants in Si and Ge. Townsend[10] found similar results based on oscillations due to interference in the transmitted intensity as a function of far-infrared frequency.

We use an optical method of determining χ which appears to be particularly suitable for investigating a phase transition at $T = 0$ K. We measure the absorption coefficient $\alpha(\omega)$ at energies $\hbar\omega$ in the range 2.5 to 58 meV and a temperature $T \approx 2$ K at which $\hbar\omega \gg kT$ and $\alpha(\omega)$ is in the zero-T limit. To obtain $\alpha(\omega)$ we pass far-infrared radiation through a Michelson interferometer, a cold filter that cuts off photons above $\omega \approx 500$ cm^{-1}, and the Si:P sample. We detect the transmitted intensity using a Ge:In bolometer at T near that of the sample, Fourier transform the resulting interferograms, and then ratio the Si:P data to a Si reference. Finally, we transform $\alpha(\omega)$ to obtain the $\omega = 0$ refractive index $n(0)$ using the Kramers-Kronig relation,

$$n(0) = n_{\text{Si}} + (c/\pi)\int_0^{\omega_\infty} [\alpha(\omega)/\omega^2]\,d\omega, \qquad (1)$$

where c is the speed of light. We correct the values of α for $n_D > 10^{18}$ cm^{-3} for the donor reflectivity using the Kramers-Kronig relations at finite frequency in a self-consistent manner. The