JOURNAL OF APPLIED PHYSICS **99**, 08P703 (2006)

Evolution of the magnetic properties and magnetic structures along the $R_mM\text{In}_{3m+2}$ (R=Ce, Nd, Gd, Tb; M=Rh, Ir; and m=1,2) series of intermetallic compounds

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(Presented on 2 November 2005; published online 28 April 2006)

We discuss the evolution of the magnetic properties and magnetic structures along the series of intermetallic compounds $R_mM\text{In}_{3m+2}$ (R=Ce, Nd, Gd, Tb; M=Rh, Ir; and m=1,2). The m=1,2 are, respectively, the single layer and bilayer tetragonal derivatives of their cubic $R\text{In}_3$ relatives. Using a mean field model including an isotropic first-neighbors Ruderman-Kittel-Kasuya-Yoshida interaction (K) and the tetragonal crystalline electrical field (CEF), we demonstrated that, for realistic values of K and CEF parameters, one can qualitatively describe the direction of the ordered moments and the behavior of the ordering temperature for these series. The particular case, where the rare-earth ordered moments lie in the ab plane or are tilted from the c axis and T_N can be reduced by tuning the CEF parameters, revealed an interesting kind of frustration that may be relevant to the physical properties of complex classes of materials such as the $R_mM\text{In}_{3m+2}$ (M=Rh, Ir, and Co; m=1,2) heavy-fermion superconductors. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2176109]

The occurrence of unconventional superconductivity (USC) in layered compounds is an intriguing phenomenon of the relationship between electronic behavior and crystal structure that is verified in many classes of USC such as the high-T_c superconductors (HTSCs), organics, and heavy-fermion superconductors (HFSs). In the latest, the recently discovered[1–3] family RMT_5 (the so-called 1-1-5), where M=Rh, Ir, and Co and T=In or Ga, has allowed a remarkable opportunity to further explore the possibility of magnetically mediated superconductivity in HFSs and its relationship with dimensionality and crystal structures. This is because the 1–1-5*s* and their structurally related bilayer (2-1-8) and cubic (1-0-3) relatives host several USC (pressure induced and at ambient pressure) including Ce-based and Pu-based compounds. Among them, PuCoGa_5 possesses a relatively high T_c that has reached the value of 18 K.[4] In addition, systematic alloying studies in $\text{CeRh}_{1-x}\text{Ir}_x\text{In}_5$ Refs. 5 and 6 and $\text{PuCo}_{1-x}\text{Rh}_x\text{Ga}_5$ Ref. 7 have revealed a linear dependence between T_c and the ratio of the tetragonal lattice parameters c/a indicating that the increasing of the quasi-two-dimensional character of their crystal structure is favoring USC.

Such as the HTSCs and the organics, the HFSs are believed to be magnetically mediated superconductors.[8] For most of them, USC seems to occur at the vicinity of a magnetically ordered state and the spin fluctuations (SF) associated with that *frustrated* magnetic phase may mediate the superconducting pair formation.[8] More recently, NMR studies in PuCoGa_5 have revealed that the SC in this material is unconventional with *d*-wave symmetry and similar properties to those found in other HFSs and HTSCs, suggesting that these complex USCs may share the same pairing mechanism.[9]

In particular, for Ce-based 1-1-5*s* HFSs, the magnetic properties are associated with the their 4*f* electrons. Despite the heavy-fermion character of these compounds, evidence for localized 4*f* moment behavior has also been found in this family.[10–12] Besides, if certain structures favor USC mediated by SF, it is an important first step to understand how layered structures can affect their magnetic properties. In this regards, detailed studies of the *f*-electron magnetism along the series of rare-earth- and actinides based 1-1-5 compounds may be very elucidative.

In the case of the Ce-based 1-1-5*s*, Nd-, Tb-, and Gd-based structurally related materials have been investigated in detail[13–19] and their magnetic properties were found to mainly depend on the interplay between crystalline electrical field (CEF) effects and Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction. For instance, among the Nd-based compounds, $\text{Nd}M\text{In}_5$ and Nd_2MIn_8, M=Rh or Ir, a systematic relationship between the antiferromagnetic (AFM) ordering

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0021-8979/2006/99(8)/08P703/3/$23.00

temperature T_N and the low-T CEF splitting was found.[14] Furthermore, when comparing the magnetic properties of the tetragonal variants $R M\mathrm{In}_5$ and $R_2 M\mathrm{In}_8$ with their cubic relatives $R\mathrm{In}_3$, T_N is significantly enhanced for the R=Nd and Tb.[6,14,19] In contrast, the tetragonal CeRhIn_5 and $\mathrm{Ce_2RhIn_8}$ present a T_N a factor of 2 smaller than that for CeIn_3, whereas, for the Gd-based materials, the low temperature magnetic properties remain nearly unaltered compared to the GdIn_3.[6,14,18] In this work we revisit the general trends of the evolution of the magnetic properties and structures along the $R_m M\mathrm{In}_{3m+2}$ (R=Ce, Nd, Gd, Tb; M=Rh, Ir; and $m=1,2$) series, including the data for the recently synthesized Tb variants.[19] Using a simple mean field model including an isotropic first-neighbors interaction and the tetragonal CEF, we show how the CEF interacts with the local spin moments giving rise to the spin ordered states that may explain those seen experimentally. We have also investigated the effect of the CEF on T_N.

As a very simple approximation to the f-electron magnetism in these series, we consider the following Hamiltonian:

$$H = K\sum_{\langle i,l\rangle} J_i \cdot J_l + \sum_i B_{20}O^0_{2,i} + B_{40}O^0_{4,i} + B_{44}O^4_{4,i}, \qquad (1)$$

where $K>0$ represent an AFM interaction between nearest neighbor local spins J_i that mimic the RKKY interaction in a simple way. The last terms are the Stevens equivalent operators that describe the tetragonal CEF in terms of powers of the local moments J. For example, $O^0_{2,i}=3J^2_{z,i}-J(J+1)$ is an operator that favors an in-plane order of spins momentum ($J_z=0$) if $B_{20}>0$ or in the c direction if $B_{20}<0$. The description of the other operator can be found in Ref. 20.

We have solved the Hamiltonian Eq. (1) with a simple mean field approximation ($J_i \cdot J_j \sim J\cdot\langle J\rangle$) but taking full account of the on-site CEF. With this approximation our Hamiltonian can be written as $zKJ\cdot\langle J\rangle$, where z is the number of nearest neighbors. In this work we have fixed zK $=1.3$ meV, which is about the order of the T_N for the cubic NdIn_3 and CeIn_3. All energy values are expressed in meV. We characterize the ground state of the Hamiltonian for a given set of CEF parameters B with the mean direction of the spin $\langle J\rangle$. We also extracted T_N, above which the Néel order is lost ($\langle J\rangle=0$), the magnetic susceptibility $\chi(T)$ and the spin fluctuations $\langle Jz^2\rangle$(TN) on the c-axis. In particular, we have chosen this last magnitude as tuning parameter.

Figure 1 summarizes the evolution of the magnetic properties along the $R_m M\mathrm{In}_{3m+2}$ (R=Ce, Nd, Gd, Tb; M=Rh, Ir; and $m=1,2$) by showing the behavior of T_N and the paramagnetic Curie-Weiss temperatures for the homologous m $=1,2$ compounds compared to their cubic relatives. As one can see, θ_p shows little change among these series for all R. This result indicates that in the molecular field approximation, the effective exchange parameter between rare earths remains about the same at high T through these homologous series. On the other hand, T_N shows significant evolution for the non-S R=Ce, Nd, and Tb materials, suggesting again that the low temperature CEF scheme configuration play a fundamental role in the observed trends. Furthermore, for compounds whose resolved magnetic structures have revealed that the R moments point along the c axis (RRhIn_5, R=Nd and Tb),[15,19] T_N is enhanced for the tetragonal variants. In contrast, T_N is suppressed to less than 0.5 of the CeIn_3 value for CeRhIn_5 and $\mathrm{Ce_2RhIn_8}$ where the Ce magnetic moments are aligned out of the c axis.

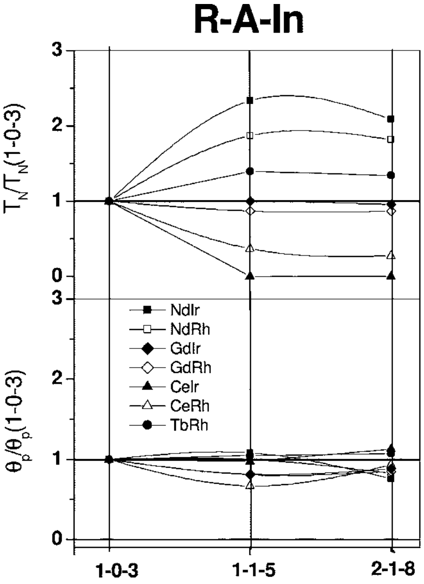

FIG. 1. Evolution of the normalized Néel and paramagnetic Curie-Weiss temperatures for the studied $R_m M\mathrm{In}_{3m+2}$ (R=Ce, Nd, Gd, Tb; M=Rh, Ir; and $m=1,2$) compounds.

In Fig. 2 we show, for zero temperature, the angle θ of the ordered moment with respect to the ab plane as a function of B_{20} varies. When $B_{20}=0$ and $B_{44}=5B_{40}>0$ the CEF corresponds to that of a cubic symmetry where the local moments tend to point in the [111] direction which corresponds to $\theta/\pi=0.2$. Naively the decrease of B_{20} (to negative values) should be associated with a tendency to order in the c direction, as discussed previously. This is, in fact, the case for $J=9/2$ (Nd^{3+} case) and $J=6$ (Tb^{3+} case) where the spin moves towards the c axis as the magnitude of B_{20} increases. But for a smaller magnetic momentum, as in Ce $J=5/2$, the quantum nature of the spin gives rise to a nontrivial evolution and the spin actually moves to the plane for small (negative) values of B_{20}. For large enough values of B_{20} the natural trend is recovered and the spin points in the c-axis direction. It is interesting to note that these very different behaviors

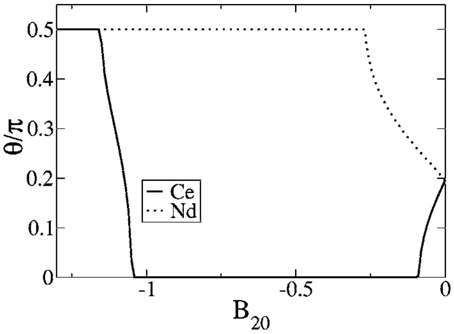

FIG. 2. Angle θ/π of the local spin with respect to the ab plane as B_{20} varies for $J=5/2$ (Ce-like case, continuous line) and $J=9/2$ (Nd-like case, dotted line). The other CEF parameters correspond to a cubic symmetry with $B_{40}=0.05$ meV.

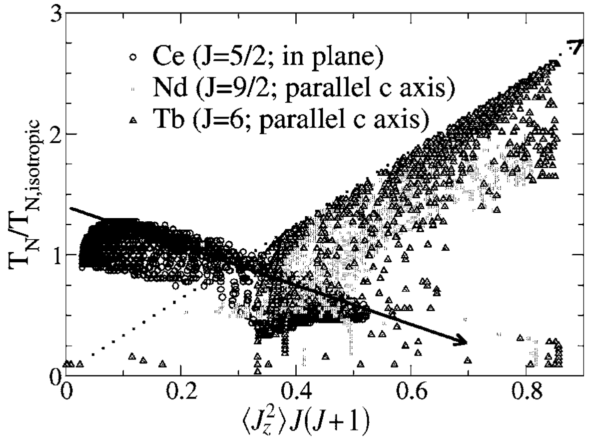

FIG. 3. Normalized Neel temperature $[T_{N,\mathrm{isotropic}}=KJ(J+1)/3]$ as a function of $\langle J_z^2 \rangle$(TN) for various CEF parameters. In the case of $J=5/2$ the data show CEF parameters for which the magnetic moment lies in the *ab* plane, while for the others spins the magnetization is parallel to the *c* axis. The arrow only shows the data trends.

happen in a parameter region close to that obtained experimentally for the Ce-115 compounds [$B_{20} \sim 1$ meV, Ref. 21 and cubic parameters close to the ones used in Fig. 2]. These simple calculations revealed that the CEF effects alone may account for the different direction of the spin AFM order in these series.

Regarding the influence of a given CEF scheme in T_N, it is a reasonably hand-waving argument that if a system orders in a given direction and we change the CEF parameters in order to make it more magnetically susceptible in some other direction, but without actually changing the order, the system may experiment some kind of magnetic frustration or the energy barrier between these states should diminish and therefore T_N should decrease as well. Inversely, if the system orders in a certain direction and we change the Hamiltonian parameters in order to favor even more this state, the ordering temperature should increase. How to explore the relation between the "tendency" to order and the actual T_N in a more precise way? We take the magnetic fluctuations on the c-axis $\langle J_z^2 \rangle$(TN) at the transition temperature as an effective measure of the tendency of the system to be in the *c* direction, even if its actual zero temperature order is in same other direction.

In Fig. 3, we show results of T_N vs $\langle J_z^2 \rangle$(TN) for a scan of CEF parameters in the region $|B_{20}| < 1.5$ meV, $|B_{40}| < 0.1$ meV, and $|B_{44}| < 0.5$ meV. For $J=5/2$ and when the system spin is on the plane (as for $\mathrm{CeRhIn_5}$) T_N decreases as $\langle J_z^2 \rangle$(TN) increases, in agreement with the thumbs rule previously stated. Also the model results in the case of a *c*-axis ordering also follow the rule, as T_N increases as $\langle J_z^2 \rangle$(TN) increases.

The enhancement of T_N along tetragonal materials been already already verified for a few rare-earth series[22–24] and our model is suggesting that it also applies to R=Nd and Tb 1-1-5*s* and 2–1-8*s* materials.

The results of Fig. 3 also show that the ordering temperature can be significantly changed (approximately up to a factor of 2) when the spins interact with a crystal field. Since for all non-*S* *R*-based 1–1-5*s* and 2-1-8*s*, the paramagnetic spin system is more susceptible with the field applied along *c* axis,[1–3,14,16] the trends observed in Fig. 3 reproduced the experimental behavior shown in Fig. 1. For the Ce-based materials where the magnetic ordered moments are not aligned along the *c* axis,[17,19] T_N is suppressed with the increasing of low-T g_c for the tetragonal compounds. Further, for R=Nd and Tb tetragonal materials whose the ordered moments point along the *c*-axis materials,[17,19] T_N is significantly enhanced when compared to that for their cubic $R\mathrm{In_3}$. Lastly, for the Gd-based materials, where the CEF effects are small the low temperature magnetic properties remain nearly unaltered compared to $\mathrm{GdIn_3}$.[6,14,18]

We have demonstrated that a mean field model including an isotropic first-neighbors magnetic interaction and the tetragonal CEF can qualitatively describe the direction of the ordered moments and the behavior of the ordering temperature for these series for realistic values of K and CEF parameters. Next step would be to evaluate to what extent this simple model can reproduce the particular behavior of each series of rare earth [e.g., the departure of De Gennes scaling for R=Ce and Nd (Refs. 13 and 14)] using CEF parameters determined experimentally. The particular case, where the rare-earth moments ordered out of the *c* axis and the T can be reduced by tuning the CEF parameters, revealed a frustration mechanism that may play some role in low-dimensional SF relevant to the physical properties of complex classes of materials such as the $R_m M \mathrm{In}_{3m+2}$ (M=Rh, Ir, and Co; $m=1,2$) HFSs. We believe that if a link between crystal structure and USC exists via local magnetism, *f-s* hybridization and SF, these relationships would shed light on why some crystal structures appear to *favor* USC, especially for magnetically mediated superconductors.

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