Neutron-diffraction study of PuAs and PuSb

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The first neutron scattering experiments on PuAs and PuSb single crystals are reported. PuAs is found to be a ferromagnet below $T_c = 123 \pm 1$ K. PuSb develops below $T_N = 85 \pm 1$ K an incommensurate ordering and undergoes, at $T_{IC} = 67$ K, a first-order transition to a commensurate phase, ferromagnetic in nature. In both compounds, moments are perpendicular to the ferromagnetic (001) planes, their low-temperature value is $\mu = (0.75 \pm 0.1)\mu_B$, indicating a Γ_8-type ground state within the Russell-Saunders level scheme. The magnetic phase diagram of PuSb has been determined. The incommensurate phase is suppressed by a small field of 8.5 kOe.

I. INTRODUCTION

The lanthanide ($4f$) and the actinide ($5f$) elements form with the pnictogen or chalcogen elements equiatomic compounds which crystallize in the simple NaCl-type structure. This family of compounds has been extensively studied,[1] and during the last few years attention has been focused on the cerium and uranium monopnictides, which have been found to exhibit very unusual magnetic properties.[2–5] The main characteristic is the existence of very anisotropic interactions, the origin of which is the mixing of f electrons with band electrons. One must emphasize the importance of neutron scattering experiments in the discovery of these unusual properties by producing unique information not only on the nature of the magnetic ordering, the phase transitions, the magnetic phase diagrams, but also on the anisotropic critical scattering and the unusual magnetic excitation spectra. These important results have been achieved because large and good-quality single crystals of cerium and uranium monopnictides have been prepared.[6] Actually, neutron experiments combined with bulk measurements, especially magnetization and photoemission experiments, have brought valuable information which has stimulated many theoretical studies. In particular, Thayamballi and Cooper[7] have pointed out the importance of the $5f$-electron hybridization for the understanding of the quite unusual properties of monopnictides. However, a good theoretical description of uranium in such compounds is rather difficult because it is intermediate between a band and a localized description.

So, it appears highly desirable to study transuranium compounds because neptunium, and more likely plutonium, is better described with a localized model for the $5f$ electrons, and the mixing with band electrons may be introduced later.

A program to grow large single crystals of neptunium and plutonium monopnictides and monochalcogenides has therefore been started. This program became possible because of the collaboration between the European Institute for Transuranium Elements (Karlsruhe), which provided unique glove-box and handling facilities, and the Eidgenössische Technische Hochschule (Zurich) which brought their crystal growth experience previously acquired for preparing uranium compounds. This collaboration has succeeded in growing the first single crystals of plutonium monopnictides.[8]

Magnetization measurements on these plutonium monopnictide single crystals of PuAs, PuSb, and PuBi (Refs. 9 and 10) indicate that PuAs and PuSb are ferromagnetic at low temperatures while PuBi is an antiferromagnet. For the three compounds the magnetization is strongly anisotropic with an easy direction along a $\langle 001 \rangle$ axis, a behavior very similar to that found in cerium monopnictides.

In this paper, we report the first neutron scattering measurements performed on plutonium single crystals of PuAs and PuSb. After a brief description of the experimental conditions we will report successively the results obtained on the ferromagnet PuAs and on the magnetic phase diagram of PuSb. These experimental results will be discussed in the last section.

II. EXPERIMENTAL SETUP

The neutron-diffraction experiments have been performed at the reactor Siloe at the Centre d'Etudes Nucléaires de Grenoble using a double-axis spectrometer equipped with a detector that can be elevated out of the scattering plane. This feature is particularly useful in examining highly anisotropic systems. The measurements were carried out with neutrons of wavelength $\lambda = 1.54$ Å provided by a graphite monochromator and filtered by pyrolitic graphite giving a $\lambda/2$ contamination smaller than 3×10^{-4}.

The samples used were good-quality single crystals of size $1\times1.7\times1.7$ mm^3 for PuAs and $1\times1.5\times2.5$ mm^3 for PuSb. They were oriented and encapsulated in aluminum containers in a glove box at the Institute for Transuranium Elements. The samples were contained in a variable temperature cryostat with a [001] axis vertical. To investigate the magnetic phase diagram of PuSb a cryomagnet was used. These crystals were grown from the isotope ^{239}Pu which has a high capture (269 barns) and fission (742.5 barns) cross sections at 25.3 meV, but with such small samples and because we concentrated on elastic scattering the absorption was not prohibitive.

III. PuAs

PuAs is found to develop a ferromagnetic ordering below $T_c=123\pm1$ K (Fig. 1). Above T_c scans along the symmetry directions of the face-centered-cubic Brillouin zone give no evidence for any superlattice peak indicating that PuAs is a true ferromagnet. Below T_c the strong nuclear reflections with even indexes $\langle200\rangle$ and $\langle220\rangle$ increase by about 25% in intensity which is too much to be accounted for by the magnetic contribution. This change in intensity can be explained only by the variation of the extinction phenomena which become less important in the ferromagnetic state because of the small size of the magnetic domains and probably due to a small tetragonal lattice distortion.[11] Although the extinction effects are important for strong reflections (larger than 20%), they can be neglected for the weak reflections with odd indexes as $\langle111\rangle$ and $\langle311\rangle$ because their intensities are more than a factor 20 smaller. Therefore only the intensities of the $\langle111\rangle$ and $\langle311\rangle$ reflections have been used to get the ferromagnetic moment. In order to avoid the problems arising from the large absorption of ^{239}Pu, the ratio between the intensities of the low temperature and the paramagnetic states have been measured from which we can determine the experimental ratio I_M/I_N between the magnetic and the nuclear intensities. Then the magnetic structure factor $F_M(\vec{h})$ is given by

$$F_M^2(\vec{h})=\tfrac{2}{3}(0.27)^2[\mu f(\vec{h})]^2=F_N^2(\vec{h})\left[\frac{I_M}{I_N}\right]_{\text{expt}},$$

where μ is the magnetic moment, $f(\vec{h})$ the $5f$ form factor, the factor $\frac{2}{3}$ takes account for the ferromagnetic domains, and $F_N(\vec{h})$ is the nuclear structure factor. By this procedure the main difficulty is to have a good determination of the scattering lengths as

$$F_N(\vec{h})=\bar{b}_{\text{Pu}}-b_{\text{As}}\ .$$

The scattering length $b_{\text{As}}=0.564\times10^{-12}$ cm is well determined, but this is not the case of $\bar{b}_{\text{Pu}}$ because it depends on the value of $b_{^{239}\text{Pu}}$, which is not known accurately. The isotope composition (90 at.% ^{239}Pu) is known, so the experimental value of interest is $\bar{b}_{\text{Pu}}$. For our sample we used the value $\bar{b}_{\text{Pu}}=0.82\times10^{-12}$ cm, slightly larger than the value determined by polarized neutron experiments on PuSb [$\bar{b}_{\text{Pu}}=0.81\times10^{-12}$ cm at $\lambda=1.1$ Å (Ref. 12)] to take into account the small wavelength dependence of the scattering length ($\bar{b}_{\text{Pu}}=0.82\times10^{-12}$ cm at $\lambda=1.54$ Å). An error of 1% in the scattering length of the Pu atom gives rise to an error of 5% in the value of the magnetic moment. The Debye-Waller factors were not taken into account because they introduce a correction smaller than 1% which is beyond the experimental accuracy.

The values 1.35 ± 0.10 and 1.32 ± 0.10 have been measured at $T=5$ K for the ratio I_M/I_N of the $\langle111\rangle$ and $\langle311\rangle$ reflections, respectively. From these values we deduce

$$\mu f(\langle111\rangle)=\mu f(\langle311\rangle)=(0.85\pm0.05)\mu_B\ .$$

The fact that the form-factor values of the $\langle111\rangle$ ($\sin\theta/\lambda=0.15$ Å^{-1}) and $\langle311\rangle$ ($\sin\theta/\lambda=0.28$ Å^{-1}) reflections are equal is in agreement with a Pu^{3+} form factor. Actually, for a Pu^{3+} ion ($5f^5$, $S=\frac{5}{2}$, $L=5$) the ground-state multiplet corresponds to an antiparallel coupling of the spin and orbital moments, and since these moments have a different extension in space, the resultant form factor exhibits a maximum at a finite value of about $\sin\theta/\lambda=0.25$ Å^{-1} as shown by polarized neutron experiments.[12,13] Therefore it is rather difficult to get an accurate magnetic moment value because the extrapolation at zero is not easy, and is somewhat dependent on the model used. Taking into account this difficulty and the uncertainty on the Pu scattering length, we estimate the moment value at $T=5$ K to be $\mu=(0.75\pm0.10)\mu_B$. This value is slightly larger than the value of $0.67\mu_B$ (Ref. 9) determined by magnetization experiments, although within statistics.

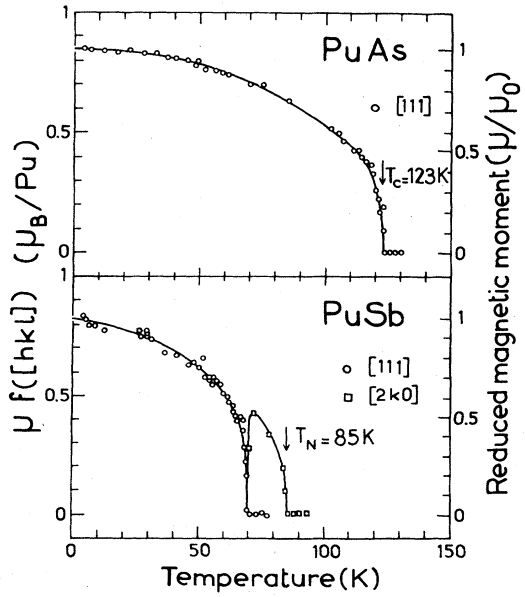

FIG. 1. Thermal variation of μf for the $\langle111\rangle$ reflection corresponding to PuAs and PuSb.

IV. PuSb

In zero applied magnetic field PuSb develops a magnetic ordering below $T_N=85\pm1$ K. Scans performed along the various symmetry directions of the reciprocal lattice

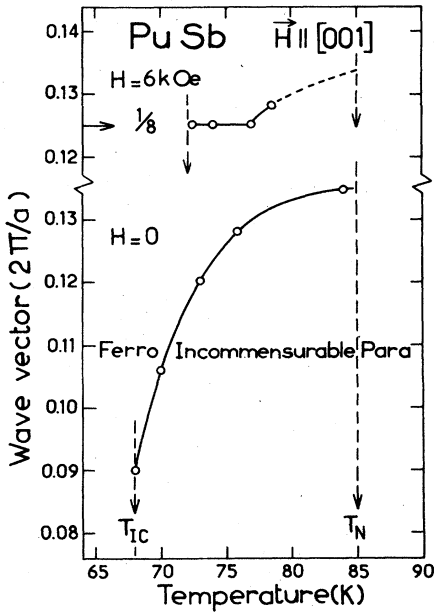

FIG. 2. Thermal variation of the value of the wave vector associated with the incommensurate phase of PuSb with and without an applied magnetic field.

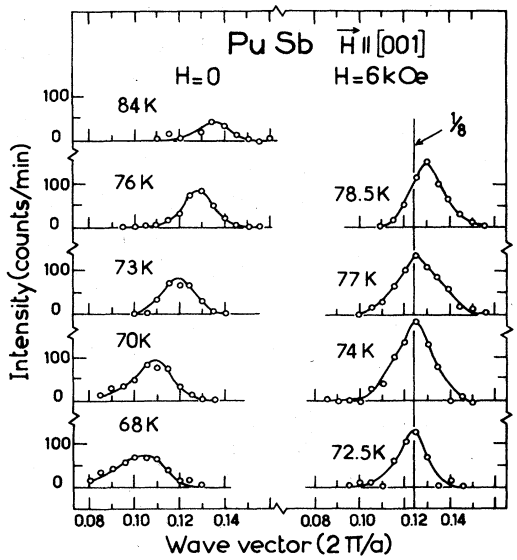

FIG. 3. Scan performed in the [2*k*0] direction showing the temperature and field dependence of the superlattice magnetic peak associated with the incommensurate phase of PuSb.

allow us to locate the magnetic scattering vectors ($\vec{h}$). They are found to be associated with a wave vector $\vec{k}=\langle 00k\rangle$ ($\vec{h}=\vec{H}+\vec{k}$, $\vec{H}$ being a Brillouin zone center); the absence of the $\langle 00k\rangle$ magnetic peak clearly indicates a longitudinal polarization of magnetic moments, i.e., the Fourier component $\vec{m}_{\vec{k}}$ is parallel to the wave vector $\vec{k}$. Such a situation is quite similar to that found in cerium and uranium monopnictides. However, in the case of PuSb the wave-vector value is incommensurate with the lattice; when the temperature is decreased it varies continuously from $k=0.135\pm0.005$ at T_N down to $k=0.090\pm0.005$ at $T_{IC}=67$ K (see Fig. 2). At T_{IC}, the temperature of the incommensurate-to-commensurate transition, a first-order transition occurs to a ferromagnetic state. The continous variation of the value of the wave vector clearly establishes the incommensurate nature of the magnetic ordering. Then PuSb behaves, in some sense, differently from cerium or uranium monopnictides which always exhibit commensurate magnetic structures.

The temperature dependence of the wave-vector value follows closely the following relation:

$$k(T_N)-k(T)=a(T_N-T)^3$$

with $a=1.25\times10^{-3}$ r.l.u./K, where 1 r.l.u. is 1 reciprocal-lattice unit. Close to T_{IC} the superlattice peaks have a larger width, indicating a distribution in the value of the wave vector which may correspond to the incomplete devil's staircase behavior.[14]

In order to characterize the incommensurate phase a careful search for the presence of second- and third-order harmonics has been undertaken, but no intensity was detected. Taking into account the experimental accuracy, which is actually not very good because of the absorption and the small size of the crystal, we can estimate that the amplitude of higher harmonics is less than one tenth of the fundamental component. Therefore, in zero field, the incommensurate state corresponds nearly to a sine-wave modulation of the magnetic moment. The magnetic ordering consists of a stacking of ferromagnetic (001) planes, with magnetic moments perpendicular to these planes and varying from plane to plane. The magnetic moment value is modulated according to a sine-wave modulation, i.e.,

$$\vec{m}(\vec{R}_n)=A_k\hat{u}_k\cos(2\pi\vec{k}\cdot\vec{R}_n+\phi_k)\ ,$$

where A_k and $\hat{u}_k$ are, respectively, the amplitude and a unit vector parallel to the wave vector $\vec{k}=\langle 00k\rangle$; ϕ_k is a phase. Thus

$$\vec{m}_k=\frac{A_k}{2}\hat{u}_k e^{i\phi_k}\ .$$

A more accurate description of the incommensurate phase and of the *I-C* transition requires a larger crystal with a plutonium isotope more suitable for neutron scattering, such as ^{242}Pu.

A magnetic field along a [001] axis strongly modifies the incommensurate phase as can be seen in Fig. 3. In the presence of a magnetic field the value of the wave vector no longer has a continuous dependence on temperature, but locks into commensurate values. As an example, for $H=6$ kOe (Figs. 2 and 3), the commensurate value $k=\frac{1}{8}$ is retained over a rather large temperature range. From the thermal variation of the magnetic intensities, associated with the incommensurate and the ferromagnetic structures (see Fig. 4), we deduce that the ordering temperature is not affected by the applied magnetic field whereas the *I-C* transition is strongly shifted to higher temperatures. A magnetic field larger than 8.5 kOe is enough to

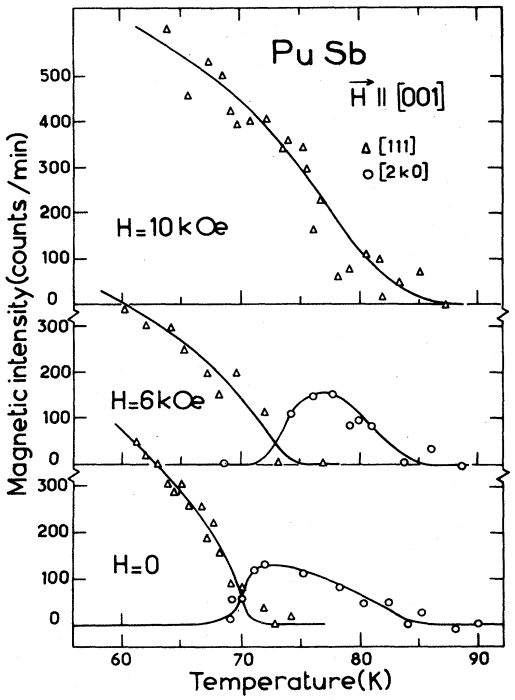

FIG. 4. Thermal variation of the magnetic intensities of PuSb with different applied magnetic fields.

suppress the incommensurate state, as can be seen in Fig. 5 which represents the (H,T) magnetic phase diagram. A better description of these commensurate phases also requires the nonabsorbing isotope.

Below the first-order transition at T_{IC}, the ferromagnetic component increases continuously with decreasing temperature to reach a saturated value below $T=5$ K. As is found for PuAs, at $T=4.2$ K, the values 0.52 ± 0.05 and 0.64 ± 0.05 have been measured for the ratio I_M/I_N of the $\langle 111\rangle$ and $\langle 311\rangle$ reflections, respectively. In PuSb we determine a slightly larger value of $\mu f(\langle 311\rangle)=(0.92\pm0.05)\mu_B$ than that of $\mu f(\langle 111\rangle)=(0.83\pm0.05)\mu_B$. Although this difference can be accounted for by the experimental uncertainties, it may indicate that the maximum of the form factor is more pronounced in PuSb than in PuAs. With the same difficulties as for PuAs we deduce a magnetic moment $\mu=(0.74\pm0.05)\mu_B$ for Pu^{3+} in PuSb. This value is indeed similar to that found for PuAs and close to the magnetization result $(0.65\mu_B)$.[9,10]

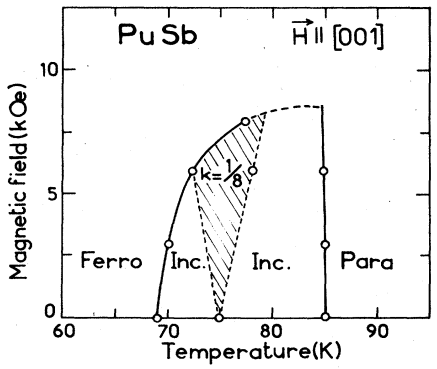

FIG. 5. (H,T) magnetic phase diagram of PuSb for a magnetic field applied along a $\langle 001\rangle$ direction.

In contrast to PuAs, in PuSb the strong nuclear reflections are not affected by the appearance of the ferromagnetic order, indicating that the extinction effects must be very small.

V. DISCUSSION

These first neutron-diffraction experiments have proved that studies of small single crystals (a few mm^3) of ^{239}Pu compounds are possible.

PuAs is found to be a ferromagnet over the whole temperature range with an ordering temperature $T_c=123\pm1$ K, in good agreement with magnetization experiments,[9] which indicate a large anisotropy with magnetic moments along a $\langle 001\rangle$ easy axis.

PuSb is not a simple ferromagnet. It orders with an incommensurate structure at $T_N=85\pm1$ K, and at about $0.8T_N$ a first-order transition leads to a commensurate ferromagnetic state ($T_{IC}=67$ K). In the incommensurate phase the magnetic structure is collinear and, as in the low-temperature ferromagnetic phase, the moments are aligned along a $\langle 001\rangle$ direction. This corresponds to a stacking of (001) planes with a quasi-sine-wave modulation of the moment value from plane to plane, the wave vector of the modulation being temperature dependent. However, a magnetic field induces some lock-in with commensurate values of the wave vector of $\frac{1}{8}$. Such a behavior cannot be understood from the theory of I-C transitions based on a continuum Landau-Ginzburg functional because it needs much higher order terms. A better description of incommensurate systems is based on the so-called devil's staircase function,[14] which results from a consideration of the discrete nature of the lattice. A similar behavior has been found in thiourea[15] which undergoes also an I-C transition with a ferroelectric low temperature ordering. As far as magnetic systems are concerned, PuSb offers a unique example to investigate lock-in effects. However, such a study can be done only with a larger crystal with ^{242}Pu.

Valuable information concerning the plutonium ground state can be deduced from the neutron-diffraction and magnetization experiments. Both indicate that the moments are aligned along a $\langle 001\rangle$ direction and give, at low temperatures, a moment value of about $0.75\mu_B$. Such a value can be explained by the Russell-Saunders coupling scheme. Within this scheme Pu^{3+} is similar to Sm^{3+} with a ground-state multiplet $J=\frac{5}{2}$ ($l=5$, $S=\frac{5}{2}$, $J=L-S$). Due to the large value of the spin-orbit coupling an intermediate coupling is more appropriate. Using the calculation done by Lam and Chan[16] the value of the Landé factor is $g_J=0.343$. Such a value gives an effective paramagnetic moment $\mu_{\text{eff}}=1.01\mu_B$, in good agreement with susceptibility results[9] ($\mu_{\text{eff}}=1.0\mu_B$) and a maximum value $g_JJ=0.86\mu_B$. However, the degeneracy of the $J=\frac{5}{2}$ multiplet is split by the cubic crystalline electric field (CEF) into a Γ_7 doublet and a Γ_8 quartet. The magnetic moment associated with Γ_7 and Γ_8 being

$0.29\mu_B$ and $0.63\mu_B$, respectively, we can exclude a well isolated Γ_7 as the ground state. Since the experimental value of the magnetic moment is somewhat larger than the Γ_8 value, the ground-state wave function in the ordered state is close to a pure $|\frac{5}{2}\rangle$ state. Such a wave function implies a large mixing between Γ_8 and Γ_7 states which is possible only if the energy difference between Γ_8 and Γ_7 is not large in comparison with the exchange interaction, for example, a few hundred degrees kelvin. A more precise ground-state wave function could be deduced from the polarized neutron study.[12]

Therefore, in the paramagnetic state the CEF level scheme must be either Γ_8 as the ground state with the Γ_7 level at a few hundred degrees kelvin above or a Γ_7 ground state with Γ_8 close to it. This situation is actually very similar to that found in CeSb or CeBi and is not that which we would expect for a normal f system. As the Stevens factor β has the same sign for both Ce^{3+} and Pu^{3+} the normal contribution leads to a Γ_7 ground-state doublet well separated from the excited Γ_8 quartet. In CeSb and CeBi the anomalously small CEF splitting was explained by Kasuya's group[17] as being due to the mixing of $4f$ electrons with the valence p band. A similar situation must occur in plutonium monopnictides to explain the ground state deduced from the observed moment value and the strong anisotropy along $\langle 001\rangle$. Thayamballi and Cooper[7] have developed a theory to extend the previous calculations[18] of the exchange interactions mediated by the mixing with band electrons to the case of several f electrons, in order to apply it to actinide compounds. The theory explained quite well the large $\langle 001\rangle$ anisotropy and the strong ferromagnetic coupling within (001) planes, a situation quite similar to that obtained for cerium monopnictides.

In plutonium monopnictides we would expect, as in CeSb or CeBi, that the effective in-plane coupling (J_0) is much stronger than the interplanar ones (J_1,J_2), a situation which corresponds to the anisotropic next-nearest-neighbor Ising model.[19–21] Taking into account only the first (J_1) and the second (J_2) interplane couplings, the theory gives a ferromagnetic or an antiferromagnetic type-I state, according to whether $J_1>0$ or $J_1<0$ if $|J_2/J_1|<\frac{1}{4}$. Whereas for $|J_2/J_1|>\frac{1}{4}$ an incommensurate phase develops below T_N, in a mean-field theory the value of the wave vector at T_N is given by $\cos\pi k=-J_1/4J_2$. If the temperature is decreased, an I-C transition occurs towards a ferromagnetic ($J_1>0$) or a type-I phase ($J_1<0$) if $|J_2/J_1|<\frac{1}{2}$, otherwise the commensurate phase is the type-IA structure $(++--)$. This theory applies quite well to cerium and actinide monopnictides, in particular for the low-temperature structures which have been found to be always ferromagnetic, type-I or type-IA. In pure compounds only PuSb was found to develop an incommensurate structure, however, similar structures have been observed in USb-UTe or USb-ThSb solid solutions.[3] In these cases, a change in the conduction-electron concentration modifies the coupling between nearest-neighboring planes (J_1), and they become ferromagnetic as the number of conduction electrons increases because the p-f mixing mechanism is suppressed. In plutonium monopnictides a similar behavior occurs because PuP and PuAs are ferromagnets with high T_c ($J_1>0$); in PuSb, J_1 still has a positive value, but in PuBi, $J_1<0$; obviously in PuX, J_1 changes sign between PuSb and PuBi. A similar change of sign occurs also in neptunium monopnictides between NpAs and NpSb, whereas in the uranium monopnictides J_1 always has a negative value. To understand this variation of J_1 both with the actinide element and the pnictides two types of exchange mechanisms must compete. The first one is well-known Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction coming from the intra-atomic d-f exchange mechanism, and the second one is the exchange interaction resulting from the mixing of $5f$ electrons with band electrons. It is this latter that gives rise to highly anisotropic interactions. The RKKY interaction, being proportional to the spin, increases on going from uranium to plutonium up to curium, whereas the mixing interaction increases as the $5f$ electrons become less localized (i.e., from Pu to U) or on going from light to heavy pnictides because the p-f mixing effect becomes larger. Therefore, in the plutonium pnictides there is a competition between the two mechanisms, and the anisotropic interactions are expected to be weaker than in the uranium and neptunium compounds. This competition may explain why the antiferromagnetic compounds PuSb and PuBi order at much lower temperature than the ferromagnets PuP and PuAs.

In conclusion, plutonium monopnictides appear to be very interesting compounds with magnetic properties in some sense similar to those of cerium monopnictides but also with significant differences. A more extensive investigation of the magnetic interactions in PuSb would provide very fruitful information. Neutron studies of the magnetic behavior of the antiferromagnet PuBi and of the neptunium monopnictides are also of great interest, and will be undertaken in the near future.

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