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**MAGNETIC TRANSITION POINTS AND MAGNETIC MOMENTS IN SOME URANIUM COMPOUNDS**

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Résumé. — Les composés polycristallins $\text{U}_3\text{P}_4$, $\text{U}_3\text{As}_4$, $\text{U}_3\text{Sb}_4$, $\text{U}_3\text{Bi}_4$ et $\text{UGa}_2$ ont été étudiés par la méthode de la dépendance $\sigma^2 = f(H/\sigma)$ pour trouver les valeurs bien définies des températures de transitions magnétiques $T_C$ et les valeurs des aimantations spontanées $(H_{\text{max}} \approx 50$ kOe, $T_{\text{min}} = 4.2$ °K). On a trouvé que les points $T_C$ coïncidaient avec les points asymptotiques de transition sauf pour les composés $\text{U}_3\text{As}_4$ et $\text{U}_3\text{Sb}_4$. On a essayé d’expliquer la différence entre les moments atomiques obtenus en états magnétiques ordonnés et désordonnés.

Abstract. — Polycrystalline compounds $\text{U}_3\text{P}_4$, $\text{U}_3\text{As}_4$, $\text{U}_3\text{Sb}_4$, $\text{U}_3\text{Bi}_4$ and $\text{UGa}_2$ have been investigated by the method of the dependence $\sigma^2 = f(H/\sigma)$ to find well defined values of the magnetic transition temperatures $T_C$ and the values of the spontaneous magnetizations $(H_{\text{max}} \approx 50$ kOe, $T_{\text{min}} = 4.2$ °K). It has been found that the points $T_C$ coincide with the asymptotic transition points except the compounds $\text{U}_3\text{As}_4$ and $\text{U}_3\text{Sb}_4$. An attempt was made of explaining the difference between the atomic moments obtained in the ordered and disordered magnetic states.

As is known, direct methods for measuring the temperature dependence of magnetic moments may fail near the points of magnetic transition. Therefore, studying the magnetic properties of uranium compounds, the authors have tried to apply the method of $\sigma^2$ vs $H/\sigma$ curves.

Polycrystalline samples of $\text{U}_3\text{P}_4$, $\text{U}_3\text{As}_4$, $\text{U}_3\text{Sb}_4$, $\text{U}_3\text{Bi}_4$ and $\text{UGa}_2$ in powder form have been prepared in vacuum by direct chemical reaction between stoichiometric amounts of their components [1]. Powdered $\text{U}$ (nuclear), obtained by thermal dissociation of $\text{UH}_3$, and $\text{P}$, $\text{As}$, etc. of semiconductor purity were used. The samples were homogenized by very long annealing, set free from impurities by magnetic separation at liquid nitrogen temperature and checked by chemical analysis. Care was taken to protect the substances from the influence of the atmosphere.

As mixtures of $\text{U}_3\text{X}_4$ with $\text{UX}_2$ (and $\text{UGa}_2$ with $\text{UGa}$ or $\text{UGa}_2$) can appear the samples were quantitatively analyzed by X-ray diffraction. The following purities (in weight %) were found: 95 % for $\text{U}_3\text{As}_4$ and $\text{U}_3\text{Sb}_4$, 90 % for $\text{U}_3\text{P}_4$ and $\text{U}_3\text{Bi}_4$ and 80 % for $\text{UGa}_2$, but can be better for each compound. According to these data all magnetic quantities treated below have been corrected to represent the materials with 100 % purity.

The procedure of measurement and interpretation was based on the relation (see, e. g., Belov [2], Heyner and Kohlhass [3] for thermodynamic derivation, and Lange et al. [4] for derivation from the molecular field model)

$$H/\sigma = a + b\sigma^2 + c\sigma^4,$$

where $\sigma$ is the magnetic moment per unit mass (assumed small in the vicinity of the transition point), $H$ the real field inside the material (high enough for complete domain alignment) and $a$, $b$, $c$ are constants (at constant temperature and pressure). Due to low magnetic moments and high fields at the experiments the external field $H$ could be used instead of $H_I$. The ballistic method of measurement has been described by the authors in [1].

Most thorough studies have been performed for $\text{U}_3\text{Sb}_4$. The experimental points of the isotherms $\sigma^2$ vs $H/\sigma$ are shown in figure 1. As predicted by eq. (1), these isotherms can be approximated by parabolas although there is a variety of forms. By full parabolas an attempt is made of keeping the vertices on a line parallel to the $H/\sigma$-axis and the coefficient $c$ from eq. (1) proportional to the absolute temperature $T$ in complete agreement with the molecular field model [4], by dashed parabolas only the condition for the vertices is fulfilled. The coincidence of full and dashed parabolas at 149.5 °K has been measured in an electromagnet, the rest in a superconducting solenoid.

![Figure 1](http://dx.doi.org/10.1051/jphyscol:19711257)
Even though a linear extrapolation from low-field measurements (i.e., up to \( \approx 10 \text{ kOe} \)) is used a deviation not more than 1 K from the value of \( T_C \) can be expected supposing a fixed temperature scale (cf. isotherm at 149.5 K in figure 1 and the results presented for classical ferromagnets by Belov [2]). Keeping this in mind we have deduced the temperatures \( T_C \) for the other compounds by the linear approximation (see Table I).

**Table I**

<table>
<thead>
<tr>
<th>Compound</th>
<th>( T_C ) (K)</th>
<th>( \Theta_C ) (K)</th>
<th>( \mu_s ) (BM)</th>
<th>( \mu_y ) (BM)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U3P4</td>
<td>144</td>
<td>145</td>
<td>-</td>
<td>2.70</td>
</tr>
<tr>
<td>U3As4</td>
<td>198</td>
<td>203</td>
<td>1.35</td>
<td>2.70</td>
</tr>
<tr>
<td>U3Sb4</td>
<td>149.5</td>
<td>154.5</td>
<td>1.55</td>
<td>3.05</td>
</tr>
<tr>
<td>U3Bi4</td>
<td>112</td>
<td>111</td>
<td>1.60</td>
<td>3.10</td>
</tr>
<tr>
<td>UGAs2</td>
<td>125.5</td>
<td>126</td>
<td>2.35</td>
<td>3.60</td>
</tr>
</tbody>
</table>

As an example figure 2 presents the temperature dependence of the spontaneous magnetization \( \sigma_s \) of \( \text{U}_3\text{Sb}_4 \) obtained both from the intersections of the isotherms \( \sigma^2 \) vs \( H/\sigma \) (Fig. 1) with the \( \sigma^2 \)-axis [3], [4] and from high-field measurements of magnetic moments at different temperatures (see below). The advantage of the former method as to the approach of the \( \sigma_s(T) \) dependence to the zero value (\( T_C \) point) is clearly seen. On the other hand, from the standpoint of general thermodynamic theory this method may be open to criticism (Fisher [5]).

In addition to the values of \( T_C \) (see above) Table I contains also the asymptotic transition points \( \Theta_C \) obtained by linear extrapolation from the dependences 1/susceptibility vs \( T \) in the region \( T > T_C \). It is seen that either the points \( T_C \) and \( \Theta_C \) coincide within the limits of experimental errors or there is a usual difference \( \Theta_C - T_C > 0 \). Thus, the differences \( \Theta_C - T_C \) reported systematically + 2 K for \( \text{U}_3\text{Bi}_4 \), \( \text{U}_3\text{Sb}_4 \) and \( \text{U}_3\text{As}_4 \) [6] or found even negative for \( \beta - \text{UH}_3 \) [7] and \( \text{U}_3\text{P}_4 \) [8], should be accepted with some caution.

Further, Table I presents magnetic moments per uranium atom (in Bohr's magnetons BM) both from the highfield (50 kOe) measurements at 4.2 K (\( \mu_s \)) and from the temperature dependence of the susceptibility above the transition point (\( \mu_y \)). Evidently, the values of \( \mu_y \) by Trzebiatowski et al. at 85 K [6], [9] are somewhat different, nevertheless, Trzebiatowski's conclusions, drawn from his values of \( \mu_y \) [9], keep their validity because of true dependence of these moments on the composition.

The relation \( \mu_y \approx 2 \mu_s \) seems to be fulfilled for our moments of cubic compounds (\( \text{U}_3\text{X}_4 \)). Using Buhler's results [10], this can be explained as follows: In a magnetically ordered single crystal each third of magnetic atomic moments \( \mu_y \) (free for \( T > T_C \)) is rigidly coupled with one of the \(< 111 > \) directions. Considering three atoms, this results in an effective magnetic moment \( \mu_y \sqrt{3} \) in the \(< 111 > \) direction, i.e., the effective magnetic moment per atom in the \(< 111 > \) direction is \( \mu_y |_{\text{eff}} = \mu_y \sqrt{3}/3 \). According to the experimental reality (extrapolation of magnetization isotherms to zero field intensities) the averaging procedure for a polycrystal means to multiply \( \mu_y |_{\text{eff}} \) by the factor \( \sqrt{3}/2 \) [11] so that for the observed \( \mu_y \) we have \( \mu_y = \mu_y |_{\text{eff}} \sqrt{3}/2 = \mu_y /2 \).

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**References**