

MAGNETIZATION DENSITY IN PARAMAGNETIC UGe_3

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by

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Abstract

We have measured the magnetization density in the actinide intermetallic compound UGe_3 (cubic $AuCu_3$ structure). The induced moment at 4.2 K with H (applied) = 48 kOe is $0.012 \mu_B/\text{mole}$. The most significant result is that a small positive moment of $\sim 0.0018 \mu_B$ is induced at and near the Ge site, which is unusual because elemental Ge is diamagnetic. The spin density at the Ge site is highly aspherical, showing the nature of the bonding with the uranium wave functions.

Introduction

The role of the outer electrons in bonding is one of particular interest in actinide compounds. In an earlier study of URh_3 (cubic $AuCu_3$ structure $a = 3.988 \text{ \AA}$) we showed [1] that the atomic susceptibility of Rh was almost doubled in this compound as compared to Rh metal. In a qualitative sense this was in agreement with the de Haas van Alphen experiments and band structure calculations [2], which suggest that a degree of electron transfer takes place from the U $6d$ to the $4d$ band. Detailed comparisons were, however, difficult because of the high degree of polarizability of the $4d$ electrons of Rh. One could argue, for example, that the increase in χ_{Rh} comes simply from an enhanced exchange interaction, rather than a direct electron transfer. dHVA experiments and calculations have suggested [3]

that a similar electron transfer and strong hybridization between the U $6d$ and $5f$ and Ge $4p$ states takes place in UGe_3 . Since elemental Ge is diamagnetic and no spin transfer has ever been reported on a Ge atom, these calculations predict something quite new and dramatic in a neutron experiment.

Experiment

The experiments have been performed on a single crystal (~ 3.5 mm diameter, 11 mm long) at the D3 polarized-neutron diffractometer at the Institut Laue Langevin, Grenoble. The neutron wavelength was 0.904 \AA and an Er filter was used to eliminate $\lambda/2$ contamination. The applied field was 48 kOe at a temperature of 4.2 K. From the susceptibility measurements the total induced moment under these conditions is $0.0124 \mu_B/\text{mole}$.

The crystal was fully characterized with unpolarized neutrons to determine accurately the nuclear structure factors, which is important because the superlattice reflections (see below) are weak and are influenced by anisotropic thermal vibrations at the Ge site even at low temperature. The extinction in UGe_3 was found to be small, unlike the case of URh_3 [1], and reliable corrections could be made easily.

The crystal structure of UGe_3 is cubic, $AuCu_3$ type with $a = 4.206 \text{ \AA}$. The structure factors fall into two groups; for hkl all even or all odd

$$N_+ = b_U \exp(-W_U) + 3 b_{Ge} \exp(-W_{Ge})$$

and for mixed hkl

$$N_- = b_U \exp(-W_U) - b_{Ge} \exp(-W_{Ge})$$

where b_U and b_{Ge} are the coherent scattering amplitudes of U and Ge, 0.852 and $0.819 (x 10^{-12} \text{ cm})$, respectively. The Debye-Waller factors are represented by $\exp(-W)$. For the magnetic structure factor M we substitute

p_i for b_i , where $p_i = 0.27 \times 10^{-12} \mu_i f_i(\vec{k})$ cm. The magnetic moment located at site i is given by μ_i , and its spatial distribution is given by the Fourier transform of the magnetization density, $f_i(\vec{k})$, where \vec{k} is the scattering vector $|\vec{k}| = 4\pi \sin \theta/\lambda$. The experiment consists of measuring the ratio of (M_i/N_i) for the Bragg reflections. Since the induced moment is small, these ratios are less than 10^{-3} , thus requiring long counting times and an appreciation of systematic errors [4]. Corrections for diamagnetism have been made in all cases [5].

Results

The experimental results are shown in Fig. 1 as a function of $\sin \theta/\lambda$. Two immediate statements can be made: (1) If all the moment is associated with the U site then a simple substitution in the equations for M_+ and M_- shows that the points from both sets of reflections should fall on a single curve. This is not the case, suggesting that $\mu_{\text{Ge}} \neq 0$. (2) The magnetic scattering is still appreciable at $\sin \theta/\lambda \approx 0.5 \text{ \AA}^{-1}$. A spin only uranium form factor falls to zero by $\sin \theta/\lambda \approx 0.4 \text{ \AA}^{-1}$ [6] so that scattering beyond this point implies the presence of an orbital moment. The analysis shows a spherically symmetric magnetization density at the U site very similar to that found in magnetically ordered uranium compounds.

To address the question of the moment localized away from the U site we have performed a Fourier inversion of the data, and this is shown in Fig. 2. Notice the spherically symmetric U magnetization density (moment $0.007 \mu_B$) and the very small and highly aspherical distribution (moment $0.0018 \mu_B/\text{Ge}$) associated with the Ge site. The significance level of the Fourier map is somewhat less than one contour in most positions and we have averaged over a small cube of 0.2 \AA on edge to reduce series termination errors.

A complete analysis of this data is still in progress. Our initial attempts to fit the Ge density with a p wave function were not successful, because the peak of the observed density is centered at about $\sim 0.6 \text{ \AA}$ from the nuclear site, whereas the $4p$ wave function has its maximum about 1 \AA from the nucleus. We speculate at this stage that the wave function has a major d component, and attempts are being made to project out the spin density at the Ge site from the band structure calculations so that a comparison can be made with experiment.

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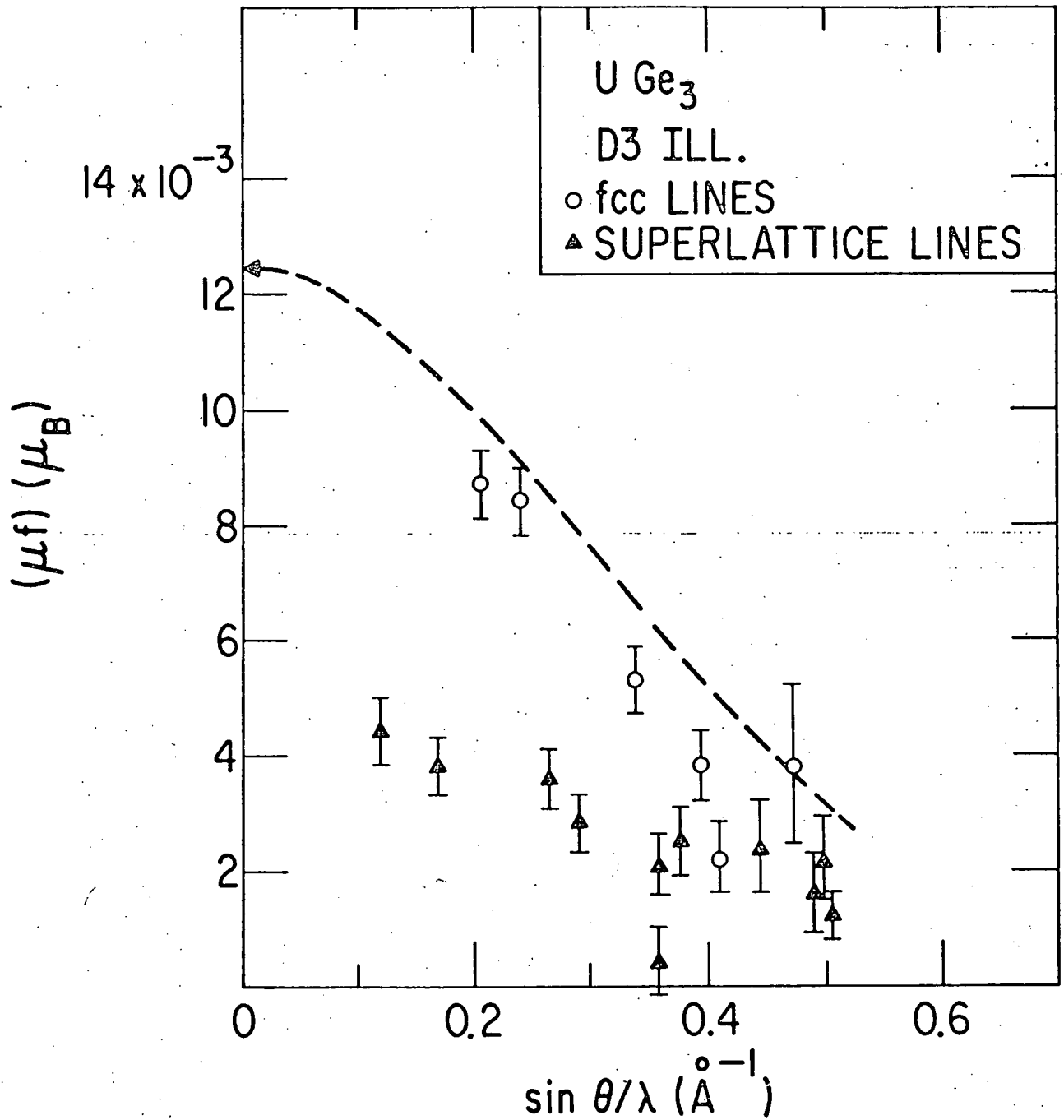
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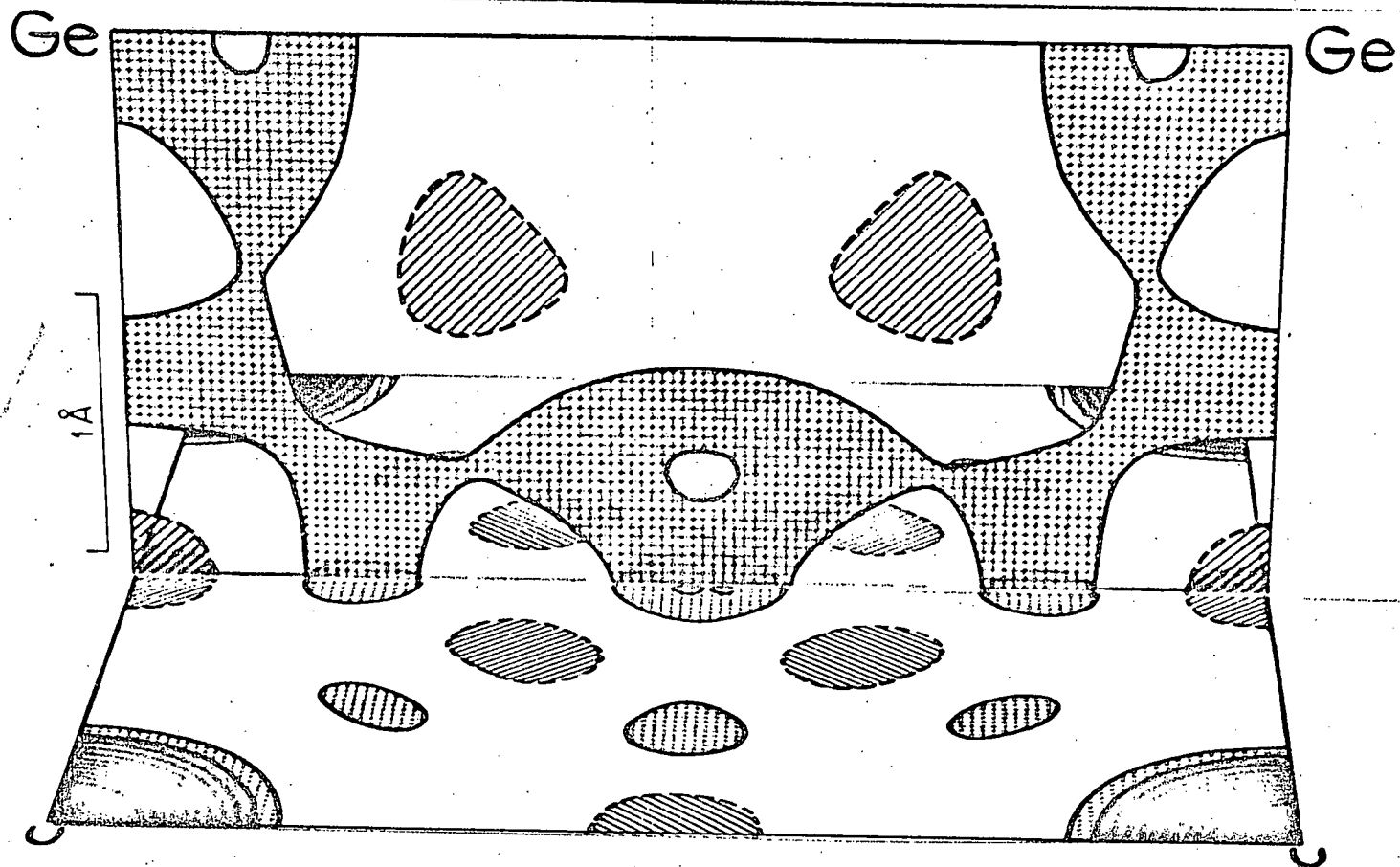
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Figure Captions

Fig. 1. Induced moment as a function of angle for UGe_3 . The dashed curve would be the form factor if all the moment was located at the uranium site. The ordinate intercept is given by the static susceptibility corrected for the diamagnetic contribution.

Fig. 2. Three dimensional picture of the magnetization density in UGe_3 . The base plane is a (001) plane showing a network of 4 U atoms. The Ge atom positions at $(\frac{1}{2}\frac{1}{2}0)$, $(\frac{1}{2}0\frac{1}{2})$, and $(0\frac{1}{2}\frac{1}{2})$ are shown. The hatched areas are positive density, the full areas being second contours (or > 2 at the U site), and the dashed areas are negative regions. The contour levels are at intervals of $1 \text{ m } \mu_B/\text{\AA}^3$ with the first contour at $-0.5 \text{ m } \mu_B/\text{\AA}^3$.





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