# Magnetic Behavior of USe at Low Temperatures\*

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Received May 25, 1971

The magnetization versus temperature behavior of USe has been investigated in the temperature range from 2.8 to 300°K and in various applied fields up to 21 kOe. A sharp decrease in magnetization is found at low temperatures. This behavior seems to be associated with the rhombohedral distortion in crystal structure, giving rise to a high anisotropy field, below the Curie temperature. Large magnetic hysteresis effects at low temperatures confirm these arguments.

# Introduction

There is at present considerable interest in the magnetic properties of uranium chalcogenides (1). Earlier investigations (2, 3) on polycrystalline USe at temperatures above 77°K have shown that this cubic compound which has the NaCl-type of structure is ferromagnetic below about 180°K. Chechernikov et al. (3) in their measurements in applied fields up to 7 kOe and temperatures above 77°K have further found that the magnetization versus temperature curves exhibited broad maxima in fields below 3 kOe. It should be mentioned that while the magnetic measurements showed the presence of magnetic ordering in USe below about 180°K, the heat capacity measurements of Takahashi and Westrum (4) indicated a lambda-type anomaly at 160.5°K. The reason for this discrepancy has not been investigated so far.

In this paper we report some new results obtained by extending the magnetic measurements down to 2.8°K and in fields ranging up to 21 kOe. The results are discussed in the light of the available information.

#### Sample Preparation and Measurements

Uranium monoselenide was prepared by a reaction between the elements which, in the powder

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form, were mixed in stoichiometric amounts, pressed into pellets in a purified argon atmosphere, encased in a tantalum tube, and then sealed in an evaculated silica capsule. The pellets were gradually heated during a period of a few days to a temperature of 800°C and then maintained at that temperature for a week. The product of the reaction was crushed, pressed once again into pellets, placed in a tantalum crucible and heated in an induction furnace to about 2000°C under vacuum for nearly 30 min. The X-ray diffraction pattern of the product material showed only a single phase with the NaCl-type structure and lattice parameter in good agreement with recent (5, 6) measurements (a = 5.740 Å).

Magnetic measurements were carried out by the Faraday method using equipment and technique now standard in this laboratory (7).

## Results

Magnetization versus temperature curves for USe are shown in Fig. 1. It is seen that at all the applied fields, there is a dramatic decrease in magnetization at the low temperature side. It should be remarked here that in all cases the sample was cooled in zero applied field and measurements were made while the sample warmed up. The maximum magnetization recorded was about  $1.0\mu_B$  per uranium atom. The inverse susceptibility versus temperature curve is also shown in Fig. 1. It yields an effective moment of  $2.4\mu_B$  per uranium atom. It should be mentioned here that there is appreciable difference between the results of Chechernikov et al. (3) and those obtained

<sup>\*</sup> This work was assisted by grants from NASA, the Gulf Oil Foundation and the Camille and Henry Dreyfus Foundations.

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FIG. 1. Magnetization (left-hand scale) at various applied fields and inverse susceptibility (right-hand scale) versus temperature for USe.

in the present study on USe in spite of the similarity in the methods of preparation. Their values for the room temperature susceptibility  $(14 \times 10^{-6} \text{ emu/g})$ and the paramagnetic moment  $(1.8\mu_B)$  are lower than our values of  $19 \times 10^{-6} \text{ emu/g}$  and  $2.4\mu_B$ , respectively. Also, our results on magnetization above 77°K are in rather poor agreement with their ones.

In Fig. 2 are shown the magnetization versus applied field curves at three different temperatures  $(2.8, 4.2, \text{ and } 78^{\circ}\text{K})$  obtained in a cycle of increasing and decreasing field strength. Large hysteresis effects are observed at 2.8 and  $4.2^{\circ}\text{K}$  while the effect at  $78^{\circ}\text{K}$  is quite small in comparison.

#### Discussion

The large decrease in magnetization at low temperatures and the sizeable hysteresis effects at 2.8 and 4.2°K seem to show the existence of a high anisotropy field in this compound at low temperatures. A very high anisotropy field of the order of 250 kOe has been estimated by Gardner (8) in the case of the isomorphous compound US. Recent low temperature crystallographic study of US and USe by Marples (6) has revealed that both of these compounds show rhombohedral distortion below their Curie temperatures. In the case of USe the interaxial angle at  $4.2^{\circ}$ K was about 89.7°, while for



FIG. 2. Magnetization versus applied field for USe at different temperatures. The arrows indicate the manner in which the field is varied.

US it was  $89.6^{\circ}$ . It has been shown by Wedgewood (9) through neutron diffraction studies that the uranium moments in US were aligned along the (111) direction and it was suggested by him that this was connected with the symmetry change to rhombohedral. The easy axis of magnetization in USe has not been determined so far, but it is quite likely to be the same as in US, since it too undergoes a rhombohedral distortion, which seems to be responsible for the high anisotropy field in USe.

It is generally believed that the uranium ion in these compounds exists in the quadrivalent state. [For some pertinent arguments favoring this viewpoint, see Grunzweig-Genossar et al. (1).] If it is assumed that the  $U^{4+}(5f^2)$  ion is predominantly in the  ${}^{3}H_{4}$  state, a pure cubic crystalline electric field will split (10) this ground state into four levels  $\Gamma_1$ ,  $\Gamma_3$ ,  $\Gamma_4$ , and  $\Gamma_5$ . In the point charge model, for an octahedral field, the singlet  $\Gamma_1$  which is nonmagnetic is expected to be the ground state. This fact may also be responsible for the low ordered moment (~1.0 $\mu_{\rm B}$ ) observed for USe in a field of 21 kOe. Although the  $1/\chi vs. T$  curve is linear in the restricted temperature range studied by us, measurements of Chechernikov et al. (3) up to about 1000°K showed marked curvature at higher temperatures. Since a detailed knowledge of the crystal field splitting is lacking at the present time, it is not possible to give a more quantitative interpretation of the observed results.

In conclusion, we believe that the low magnetization of USe at very low temperatures is due to the influence of the anisotropy field caused by rhombohedral distortion below its Curie point.

## Acknowledgment

The authors would like to express appreciation to Professor W. Trzebiatowski for drawing attention to this problem.

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