Solid State Communications, Vol. 15, pp. 583–585, 1974. Pergamon Press. Printed in Great Britain

INFLUENCE OF CRYSTAL FIELDS ON THE MAGNETIC FORM FACTOR OF SAMARIUM

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(*Received* 23 *April* 1974 *by G.W. Rathenau*)

It is calculated that crystal fields, with inclusion of mixing of higher multiplets into the ground $J = 5/2$ state, have a substantial effect on the magnetic form factor of Sm^{3+}. From the neutron diffraction data obtained by Koehler and Moon for the cubic site in Sm metal it follows that the quartet Γ_8 is lowest, with the sixth-order crystalline field parameter $A_6\langle r^6\rangle$ positive. The conduction electron moment residing on Sm is estimated to be of the order of $0.4\,\mu_B$.

RECENTLY, Koehler and Moon determined the magnetic structure of samarium metal and reported a remarkable dependence of the neutron scattering amplitudes on the scattering angle.[1,2] The scattering amplitudes of the cubic site show a rapid decrease as $\sin\theta/\lambda$ approaches zero. Moon and Koehler logically separated the magnetic form factor μf into a local $4f$ component $(\mu f)_{4f}$, which of course is affected by the crystal field splitting, and a diffuse component $(\mu f)_D$ due to spin polarization of the conduction electrons. No account was however taken of *crystal field induced admixtures of higher multiplets* into the ground $J = 5/2$ state. In the present paper we will investigate to which extent the experimental data on μf can be explained by crystal field effects and to which extent the assumption of an appreciable contribution of conduction electron spin polarization is justified. Earlier, we have shown that in the case of samarium intermetallic compounds,[3–5] in particular for $SmAl_2$ and $SmFe_2$, it is necessary to take account of an appreciable crystal field induced mixing of the nearby excited multiplets into the ground state. It also appeared that it is essential to include sixth-order potentials in the crystal field Hamiltonian. As a consequence of the crystal field induced mixing the ground state moment is in general strongly reduced.[3,5] This moment reduction leads to a more satisfactory description of the observed saturation moments of SmX_2 compounds (X = Al, Ir, Rh, Ni, Pt) than a moment reduction based on spin polarization of the conduction electrons alone.[5,6]

The scattering amplitudes discussed by Moon and Koehler are those of the cubic site in samarium metal.[2] In the crystal-field Hamiltonian we therefore restrict ourselves to terms of cubic symmetry:

$$\mathcal{H}_c = A_4 \sum_i (f_{40} + 5f_{44}) + A_6 \sum_i (f_{60} - 21 f_{64}), \quad (1)$$

in the usual notation with the summation over all $4f$ electrons. The z-axis has been chosen parallel to one of the [001] cubic axes. The calculation of the matrix elements of $\mathcal{H}_c$ has been described in previous papers.[3,4] The energy levels of the Sm^{3+} ion are then obtained by diagonalization of the Hamiltonian

$$\mathcal{H} = \lambda \mathbf{L}\cdot\mathbf{S} + \mathcal{H}_c + 2\mu_B \mathbf{H}_{ex}\cdot\mathbf{S}, \quad (2)$$

with $\mathbf{H}_{ex}$ parallel to either [001], [011], or [111], and with $\lambda/k_B = 410\,K$. In our calculations all diagonal as well as off-diagonal elements of $\mathcal{H}_c$, $\mathbf{L}$

and **S** within and between the ground state $J = 5/2$ and the lowest two excited states $J = 7/2$ and $J = 9/2$ have been considered. The expectation values $\langle L_z + 2S_z \rangle$ and $\langle S_z \rangle$ of the ground level in the exchange-split level diagram, obtained in this way as a function of the parameters $A_4\langle r^4 \rangle$, $A_6\langle r^6 \rangle$, and H_{ex}, have already been presented in a diagrammatic form in reference 5. For further details of the calculations we refer to references 3–5. The elastic neutron scattering amplitudes were subsequently calculated by use of the dipole approximation,[7]

$$(\mu f)_{4f} = (\langle j_0 \rangle + \langle j_2 \rangle)\langle L_z + 2S_z \rangle - 2\langle j_2 \rangle \langle S_z \rangle, \quad (3)$$

where we have used the values of $\langle j_0 \rangle$ and $\langle j_2 \rangle$ for Sm^{3+} tabulated by Blume, Freeman, and Watson[8] as a function of scattering angle. Since the exchange field at the cubic site in Sm is rather small ($T_C = 13.8$ K), we have restricted the computation to vanishing H_{ex}, and calculated the function $(\mu f)_{4f}$ vs the scattering angle for various sets of values of $A_4\langle r^4 \rangle$ and $A_6\langle r^6 \rangle$. The results were compared with the data given by Koehler and Moon. The following observations can be made:

(i). It appears impossible to obtain a fit to the experimental data covering the whole range of scattering angles.

(ii). A reasonable fit covering most of the experimental data can be obtained only for a limited number of sets of crystal field parameters. All these sets have the quartet Γ_8 as lowest state, with a positive sixth-order crystal field parameter $A_6\langle r^6 \rangle$. In Fig. 1 we present some of the calculated $(\mu f)_{4f}$, together with the experimental data of Moon and Koehler.[2] In order to show the dependence of $(\mu f)_{4f}$ on the sixth-order term $A_4\langle r^4 \rangle$ is kept constant and $A_6\langle r^6 \rangle$ varied between -150 and $+150$ K. It is seen that the presence of relatively large sixth-order terms can under certain conditions lead to a marked reduction in form factor at low scattering angles. This is accompanied, however, by a less good agreement with the experimental data in the region $\sin\theta/\lambda > 0.2$, where significant contributions of $(\mu f)_D$ are considered to be absent.[9] In view of the results of Davis (see reference 2) essential improvement is not to be expected in this region by using more exact calculations involving relativistic Hartree–Fock–Slater wave functions. For comparison, we have further included in Fig. 1 the free ion form factor, and the form factor for several sets of crystal-field parameters

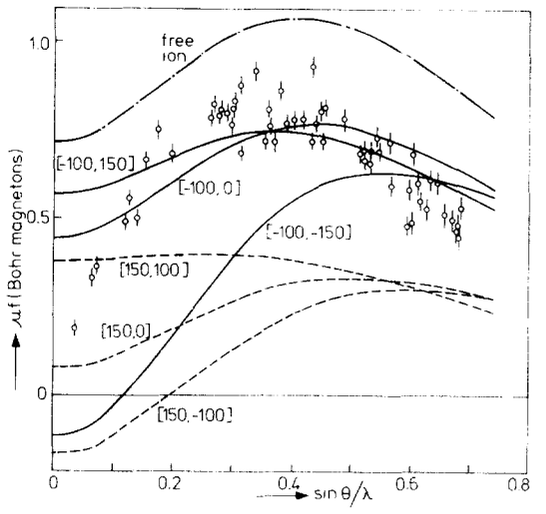

Fig. 1. Calculated magnetic scattering amplitudes $(\mu f)_{4f}$ for Sm^{3+} in the limit of small exchange $H_{ex} \to 0$, parallel to [001]. The upper curve is for free ions (zero crystal fields). The other drawn curves are for the quartet Γ_8 lowest with $A_4\langle r^4 \rangle/k_B = -100$ K and $A_6\langle r^6 \rangle/k_B = +150$, 0, and -150 K, as indicated. The dashed lines are for the doublet Γ_7 lowest with $A_4\langle r^4 \rangle/k_B = +150$ and $A_6\langle r^6 \rangle/k_B = +100$, 0, and -100 K. Experimental data for the cubic sites in Sm metal, as measured at helium temperature, are taken from reference 2.

with the doublet state Γ_7 lowest. Evidently, there is no agreement with experiment in the latter cases.

(iii). The sets of crystal-field parameters for which a reasonable fit could be obtained, correspond to an axis of easy magnetization[4] parallel to [001]. This is consistent with the experimental observation,[1] whereas a doublet Γ_7 ground state would imply the easy direction along [111].

In summary, crystal field effects are important but by themselves do not provide a complete explanation of the magnetic form factor observed for samarium metal, even if crystal-field induced mixing of higher excited states is properly taken account of, and sixth order potentials are included. Discrepancies mainly occur at low scattering angles. As already suggested by Koehler and Moon,[2] the residual decrease of the magnetic form factor for $\sin\theta/\lambda < 0.2$ is to be attributed to conduction electron polarization. From our results in Fig. 1 the conduction electron moment on Sm is then estimated to be approximately $0.4\,\mu_B$.

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Es wird berechnet, dass Kristallfelder, wobei auch das Hineinmischen von angeregten Multiplet-Zuständen in den $J = 5/2$ Grundzustand berücksichtigt wird, einen grossen Effekt auf den Formfaktor von Sm^{3+} haben. Anhand der von Koehler und Moon für die kubische Gitterstelle in Sm-Metall mittels Neutronenbeugung erziehlten Daten kann gezeigt werden, dass der Quartet-Zustand Γ_8 am tiefsten liegt wobei der Kristallfeld-Parameter 6.Ordnung $A_6\langle r^6\rangle$ positiv ist. Der Beitrag des magnetischen Moments der Leitungs-Elektronen beträgt etwa $0.4\,\mu_B$.