

Anomalous absence of effective magnetic fields at the Fe^{57} nuclei in the magnetically ordered compounds CeFe_2Si_2 and CeFeSi

E. M. Levin, R. V. Lutsiv, G. V. Popov, and S. I. Yushchuk

L'vov State University

(Submitted 31 October 1977)

Pis'ma Zh. Eksp. Teor. Fiz. **26**, No. 11, 740-742 (5 December 1977)

We investigated resonant absorption of gamma quanta in the compounds CeFe_2Si_2 and CeFeSi , which have magnetic-ordering temperatures 700 and 440 K, respectively. The character of the Mössbauer spectra demonstrates identity of the electronic states of the iron atoms and an anomalous absence of effective magnetic fields at their nuclei.

PACS numbers: 76.80.+y, 75.50.Bb

The compound CeFe_2Si_2 , as well as the series of compounds with composition RM_2Si_2 , where R is a rare-earth element and $\text{M}=\text{Cr, Mn, Fe, Co, Ni}$, crystallizes in the tetragonal structure of CeGa_2Al_2 (superstructure to BaAl_4) with space group $14/mmm$.^[1] This structure is characterized by alternation of layers perpendicular to the Z axis, of the type $\text{R}[\text{Si-M-Si}]\text{R}'[\text{Si-M-Si}]\text{R}$, where R and R' are rare-earth atoms occupying different crystallographic positions. The compound CeFeSi belongs to the group of compounds of the type RMSi , which have the structure of PbFCl (space group $P4/nmm$) and layer alternation $\text{R}[\text{Si-M-Si}]\text{R}'\text{R}$.^[2]

In both structures, each atom of the 3d element is located at the center of a tetrahedron made up of four silicon atoms, two of which belong to the upper layer and two to the lower one. In the second coordination sphere of the iron atom are located four atoms of the rare-earth element. Since the nearest environment of the iron atoms is identical in the compounds CeFe_2Si_2 and CeFeSi , it was of interest to make a comparative study of the states of their electron shells. The nuclear gamma resonance method was used for the investigation.

The spectra of absorption by the Fe^{57} nuclei in both components were measured at the temperatures 80 and 290 K. The gamma-quantum source was Co^{57m} in Pd. The polycrystalline samples of the compounds were synthesized by fusing the initial components in vacuum in alundum crucibles. The compositions of the alloys were investigated by x-ray phase analysis and x-ray-spectrum microanalysis; no extraneous phases were observed.

All the obtained spectra are characterized by a narrow absorption line at near-zero velocity (see Fig. 1). The absence of quadrupole splitting agrees with the cubic symmetry of the environment of the iron atoms in both structures. The values of the isomeric shift (relative to Fe^{57} in iron) are negative in both compounds within the limits of the measurement error (-0.05 mm/sec) and are the same at $T=80$ and 290 K. In practice, no Zeeman splitting is observed in the spectra of both compounds.

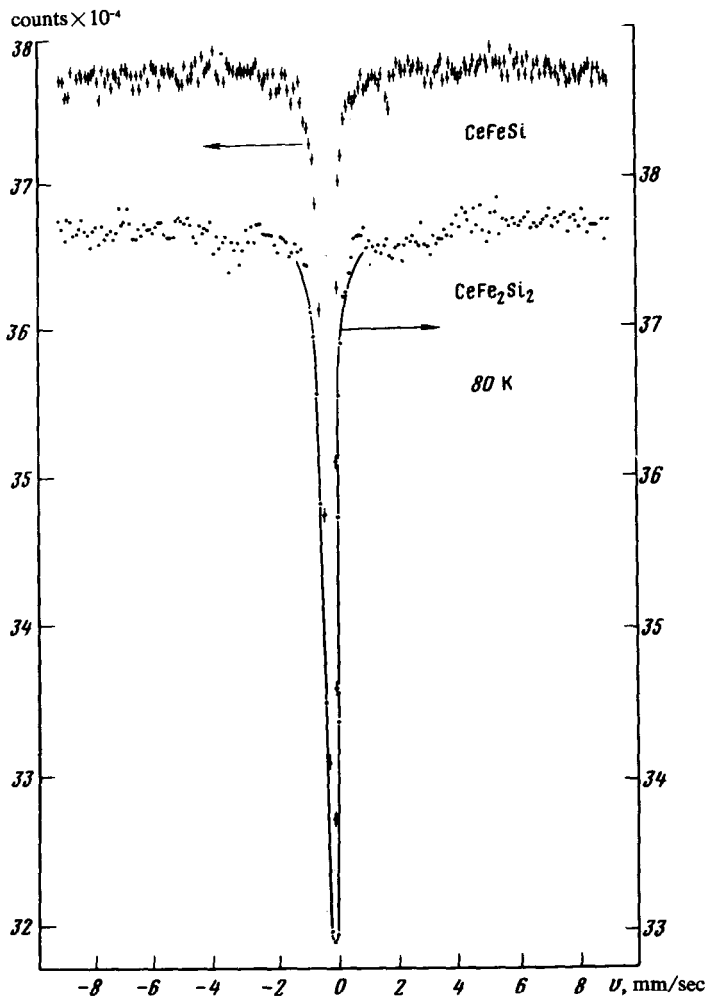


FIG. 1. Spectra of gamma-quantum absorption by Fe^{57} nuclei in the compounds CeFeSi and CeFe_2Si_2 at a temperature 80 K.

The last circumstance is unusual, since magnetic measurements have shown that CeFe_2Si_2 and CeFeSi become magnetically ordered at temperatures 700 and 440 K, respectively. We note also that investigation of a compound having the same structure as CeFe_2Si_2 , namely NdFe_2Si_2 , carried out by a neutron-diffraction method,^[3] has revealed that the iron sublattice contains a collinear spin structure of complicated character with a magnetic moment $1.5 \mu_B$ per atom. In addition, in a number of RFe_2Si_2 compounds of equal structure, the temperature of the transition to the ordered state depends on the distance $\delta_{\text{Fe-Fe}}$ in a manner that depends on the type of the rare-earth element. It is possible that the difference between the magnetic-ordering temperature

of CeFe_2Si_2 and CeFeSi is also due to the difference between the distances $\delta_{\text{Fe-Fe}}$ (2.81 and 2.87 Å, respectively). The presented data allow us to assume that the iron atoms in CeFe_2Si_2 and CeFeSi have an uncompensated magnetic moment. The existence of magnetic ordering in the compounds of the type RFe_2Si_2 and RFeSi can be attributed to the presence of exchange interactions in the Fe layers as a result of overlap of the wave functions of the 3d electrons, since the sum of the Slater radii of the 3d orbits of the iron atoms is commensurate with the distance $\delta_{\text{Fe-Fe}}$ in these compounds. Nor can we exclude the existence of indirect exchange interactions between the iron atoms via the intermediate silicon atoms, both in the layers and between them.

The absence of a Zeeman hyperfine structure in the spectra, i.e., the zero values of the effective magnetic field H_{eff} at the iron-atom nuclei, can be due to the mutual cancellation of the contributions of the s electrons to $|\psi_S(0)|^2$ and is caused by the covalent character of the bond of the iron atoms with the surrounding silicon atoms.

We have thus established that in the compounds CeFe_2Si_2 and CeFeSi , which are layered magnetically ordered crystals, the electronic states of the iron atoms are identical and that there are no effective magnetic fields at the nuclei of these atoms.

¹W. Rieger and E. Parthe, *Monatshefte für Chemie* **100**, 444 (1969).

²O.I. Bodak, E.I. Gladyshevskii, and P.I. Kripyakevich, *Zh. Strukt. Khim.* **11**, 305 (1970).

³H. Pinto and H. Shaked, *Phys. Rev. B* **7**, 3261 (1973).

⁴I. Felner *et al.*, *Solid State Commun.* **16**, 1005 (1975).