Pis'ma v ZhETF, vol. 106, iss. 9, pp. 567 – 568

Self-consistent mapping of the *ab initio* calculations to the multi-orbital p-d-model: Magnetism in α-FeSi$_2$ films as effect of local environment

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Submitted 4 October 2017

DOI: 10.7868/S0370274X17210068

In iron silicides, local magnetic moments on iron atoms strongly depend on the local environment and may be higher than in pure iron, or completely disappear when the environment changes. There are a number of experimental studies, where it was shown that one cannot neglect the role of the local environment in the formation of the magnetic state in iron silicides. However, theoretically, the influence of the local environment on the formation of the magnetic moment on iron atoms in Fe–Si alloys, on the one hand, is difficult to derive from ab initio calculations based on the density functional theory, and on the other hand, simple models often do not reflect the properties of specific compounds. Therefore, in this paper, it is proposed to combine ab initio calculations with model ones using the following scheme. First, the calculation of the electronic and magnetic properties of the compound of interest is performed within the framework of DFT-GGA for a different way of substituting silicon atoms by iron atoms or vice versa. Then, an effective multi-orbital model is constructed that takes into account all the symmetries and orbitals of the compound within the framework of the Slater–Coster approach, and the results of ab initio calculations are compared with the model ones. All details of the model calculation are given in [1]. The model parameters are selected from the requirement of matching of the charge density (as well as the density of electronic states), obtained self-consistently within the framework of GGA-DFT and within the model one. The model should help to clarify: (i) the mechanisms of local formation of the magnetic moment in iron silicides; (ii) the role that the local environment plays.

Within the framework of the proposed approach, a study was made of the mechanism of the formation of a magnetic moment in iron disilicide α-FeSi$_2$ and ordered solid solutions enriched in iron α-Fe$_x$Si$_{2-x}$ and silicon α-Fe$_{1-x}$Si$_{2+x}$. Model calculation showed that the main parameter responsible for the formation/destruction of the magnetic moment in α-FeSi$_2$ is the hopping integral t_3 between the next-nearest Fe–Fe neighbors located along the crystallographic axes of the cell. There is a sufficiently wide region of stability of the nonmagnetic state, depending only on the value of the hopping integral t_3, its decrease leads to the appearance of a ferromagnetic phase. The reason for such a critical role of the second neighbors is related with the arrangement of Fe–Fe pairs along the crystallographic axes of the cell. In this case, strong σ-bonds are formed between the d-orbitals of iron atoms, which leads to the delocalization of the d-electrons of iron atoms and the destruction of the magnetic moment. This conclusion, based on a model calculation, was confirmed ab initio by calculation and experimental data.

Our calculation within the framework of the VASP package of ordered solid solutions of α-Fe$_x$Si$_{2-x}$ with different concentrations of additional iron atoms at silicon positions showed that the appearance of one additional iron atom in the structure leads to the appearance of a large magnetic moment on it, regardless of the degree of order. However, a further increase in the number of additional iron atoms leads to the appearance of both ferromagnetic phases and nonmagnetic and ferrimagnetic phases at the same concentration of additional iron atoms, but with different spatial arrangements. The reason for the coupling of the emerging magnetic state with the spatial arrangement of the impurity iron atoms was investigated in the framework of the model approach. It was found that the main role in the appearence of a specific magnetic state is played by the number and spatial arrangement of Fe–Fe pairs in the second coordination sphere, located along the crystallographic axes. Since the different spatial arrangement of the additional iron atoms leads to different distortions of the lattice, the hopping between iron atoms in the second coordination sphere turns out to be different, which entails the appearance of different magnetic configurations. In this

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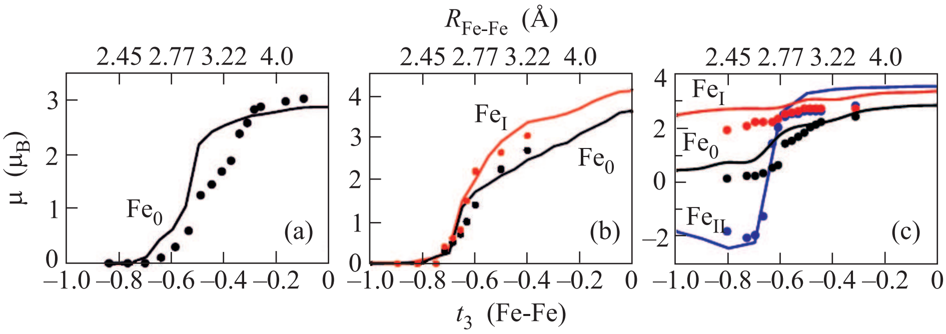

Fig. 1. (Color online) Dependence of magnetic moments on iron atoms on the hopping integral between the second Fe–Fe neighbors (lower scale) and the corresponding distances (upper scale) for different concentrations of additional iron atoms: (a) – stoichiometric α-FeSi$_2$, (b) – a structure containing three additional atoms iron, (c) – a structure containing four additional iron atoms

case, the change in the remaining hopping parameters does not play an essential role. For example, it has been obtained that when this parameter alone changes, the d-electrons are gradually delocalized and the magnetic moment decreases, and when this parameter reaches a certain critical value, a sharp transition from the ferromagnetic state to the ferromagnetic or nonmagnetic state occurs, depending on the location and concentration of the additional atoms of iron (Fig. 1).

Moreover, the increase in the number of Fe–Fe pairs in the second coordination sphere leads to the suppression of the magnetic moment and vice versa. Using the results obtained in the model calculation, it was suggested that an increase in the silicon concentration can also lead to the appearance of ferromagnetism in solid solutions of α-Fe$_{1-x}$Si$_{2+x}$. For this, a necessary condition is the change of the second coordination sphere of iron atoms by the introduction of silicon atoms, so that the number of Fe–Fe pairs becomes smaller. This assumption was verified by calculating the magnetic properties of α-Fe$_{1-x}$Si$_{2+x}$ solid solutions within the framework of the VASP package and received confirmation. Thus, an alternative explanation was proposed for the experimentally observed ferromagnetic behavior in thin films of α-FeSi$_2$, namely: the occurrence of ferromagnetism in them can be associated with an increase in the concentration of silicon atoms. Based on this statement, theoretical recommendations on growth conditions, selection of lattice parameters of substrates, and orientation of the films for obtaining a given magnetic state in α-FeSi$_2$ films are proposed.

Full text of the paper is published in JETP Letters journal. DOI: 10.1134/S0021364017210044

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