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First-principles theory of magnetically driven anomalous ordering in bcc Fe-Cr alloys

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Ab initio perturbation-theory techniques, such as the generalized perturbation method and magnetic force theorem, are used to determine the Heisenberg exchange interaction parameters and the effective cluster interactions in Fe-rich bcc Fe-Cr alloys in different magnetic states. We establish a direct connection between chemical and magnetic exchange interactions, as well as their dependence on the global magnetic state of the alloy. These findings have important implications for phase equilibria in magnetic alloys. In particular, we demonstrate that the experimentally reported concentration interval of anomalous ordering in Fe-Cr alloys is determined by the thermal history of the alloys through the value of global magnetization at the annealing temperature.

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Ferritic stainless steels based on the Fe-Cr alloy system have found numerous industrial applications during the past and the present century. Phase equilibria in these alloys have therefore been the subject of thorough experimental and theoretical investigations.[1–8] Particular attention has been paid to the spinodal decomposition of a homogeneous bcc Fe-Cr solid solution onto Fe-rich and Cr-rich fractions. This decomposition is believed to be the origin of the so-called 475 °C embrittlement,[9,10] which sets a temperature limit for applications of high-Cr ferritic and duplex steels.

A potential application of the ferritic stainless steel as a construction material for nuclear fusion reactors[11] has recently revived scientific interest to the Fe-Cr alloys. The computational physics community is again paying attention to an almost forgotten problem of the nontrivial alloying behavior in this system.[12,13] Modern computers and advanced first-principles methods open new possibilities to establish a microscopic picture of the ordering and decomposition phenomena in Fe-Cr alloys.[14–19]

The anomalous alloying behavior in the Fe-Cr system was predicted theoretically by Hennion[12] in 1983 using the tight-binding generalized perturbation method (GPM).[20] Hennion calculated the effective pair interactions in the ferromagnetic (FM) state of Fe-Cr alloys and found that the nearest-neighbor (NN) interaction was quite large and positive up to 25 at. % of Cr, indicating a tendency toward ordering (alloying) within that concentration range, while all experiments had reported only a clustering (phase separation) behavior in the Fe-Cr system. Just one year later, Mirebeau *et al.*[13] performed diffuse-neutron-scattering measurements of the atomic short-range order (ASRO) in three Fe-Cr alloys, and they indeed found ordering at the first and second coordination shells in the FM state of Fe-rich alloys, although the transition to clustering behavior was observed at a much lower Cr concentration (about 10 at. %) than predicted theoretically.

In 2003, the anomalous alloying behavior of Fe-Cr alloys was rediscovered by Olsson *et al.*[15] using first-principles calculations based on the coherent potential approximation; their results were repeatedly confirmed by supercell calculations.[15–18] All these first-principles studies dealt with calculations of the mixing enthalpy for a number of bcc Fe-Cr alloys of various compositions and atomic configurations. The mixing enthalpy in the FM state was found to change from negative to positive values upon increasing the Cr concentration beyond 6–10 at. % of Cr, depending on the state of atomic order.

These results seem to be in good agreement with the ASRO measurements by Mirebeau *et al.*[13] However, a quantitative comparison is problematic here because the sign of the mixing enthalpy and the sign of an effective pair interaction for a specific alloy composition are not directly related to each other. This is so because the mixing enthalpy is a combination of the energies of *three different systems*; one is at the composition of interest and two others are pure bcc Fe and Cr. Also, the latter two systems are in *different magnetic states*, while in the former the interaction changes with composition due to the concentration dependence of the electronic structure.

Of course, the GPM calculations by Hennion[12] were based on a simplified tight-binding model of the electronic structure, so that their results may have quite big "error bars." However, even the most recent calculations[19] of the effective pair interactions, employing a first-principles version of screened GPM,[21,22] predict that the effective pair interaction at the NN shell changes sign at a concentration of about 15 at. % of Cr, which is again substantially higher than the concentration reported experimentally.[13]

Although the remaining difference does not seem to be very dramatic (keeping in mind that first-principles calculations anyway involve a number of approximations), in this paper we demonstrate that the difference originates from a nontrivial dependence of the interatomic and effective cluster interactions (ECIs) on *temperature*. The physics underlying this behavior is currently missing in all the previous empirical and first-principles-based models of this system. At the same time, it is clear that any reliable and accurate microscopic model that aims at explaining and predicting the phase equilibria in Fe-Cr alloys should take proper account of the finite-temperature effect described below.

As will be shown below, such a nontrivial temperature dependence of the ECI originates from their dependence on the global magnetic state of the system. Let us note that the strong influence of the magnetic state on the effective cluster interactions has been demonstrated a long time ago by Staunton *et al.*,[23] who found different ordering behaviors in Ni_3Fe in the Stoner-paramagnetic and ferromagnetic states. Ling *et al.*[24,25] investigated the ordering behavior in spin-glass systems such as Cu-Mn and Au-Fe in the paramagnetic disordered local moment (DLM) state. In particular, they found the type of ordering in Au-rich Au-Fe alloys to be dependent on the magnetic state: It exhibited a $\langle 1\frac{1}{2}0\rangle$ type of ordering in the DLM state and a $\langle 100\rangle$ type in the ferromagnetic state.[25] More recently, a similar dependence of atomic ordering on the magnetic state has been demonstrated for Fe-rich Fe-Ni alloys by Crisan *et al.*[26] and Ruban *et al.*[27] In all these cases, the difference in the ordering behavior was actually observed between completely different magnetic states. In this paper, we demonstrate that even gradual changes in the global magnetization of alloys in the ferromagnetic state can influence the ECI.

We investigate the origin of the anomalous alloying behavior of Fe-rich Fe-Cr alloys using first-principles calculations of the effective cluster interactions (ECIs), including the effective pair interactions (EPIs) as well as the effective three- and four-site ECIs. The ECI are parameters of the atomic configurational Hamiltonian,

$$H_{conf}=\frac{1}{2}\sum_{p}V_{p}^{(2)}\sum_{i,j\in p}c_{i}c_{j}+\frac{1}{3}\sum_{t}V_{t}^{(3)}\sum_{i,j,k\in t}c_{i}c_{j}c_{k}+\frac{1}{4}\sum_{q}V_{q}^{(4)}\sum_{i,j,k,l\in q}c_{i}c_{j}c_{k}c_{l}. \qquad (1)$$

Here, $V_f^{(n)}$ $(f=p,t,q)$ are the n-site effective cluster interactions and c_i occupation numbers, taking on values 0 or 1 if site i is occupied by Fe or Cr atom, respectively. The summation in Eq. (1) is carried out over all sites. Indices p, t, and q designate the coordination shell for the effective pair interactions $V_p^{(2)}$ or the type of triangle and tetrahedron for the effective three- and four-site interactions, $V_t^{(3)}$ and $V_q^{(4)}$, respectively. Since in the following we will mainly deal with the effective pair interactions, we omit the upper index, $V_p \equiv V_p^{(2)}$.

The ECI can, in fact, be defined in different ways, depending on which energy is expanded (total energy, enthalpy of formation, configurational energy, and so on) and on the external constraints under which the expansion is done. Within the screened generalized perturbation method (SGPM),[21,22] they are defined at a particular volume, concentration, and magnetic state of a random alloy as an expansion of the configurational energy.[20] This is exactly what is needed for comparison with the diffuse-neutron-scattering experiments,[13] which have been made for specific compositions of bcc random Fe-Cr alloys annealed at 703 K. In general, one should also take the so-called strain-induced interactions[28] into consideration. However, a quite small atomic size mismatch between Fe and Cr allows us to assume that such interactions should be relatively unimportant in this system; they will therefore be neglected.

The electronic structure calculations for random Fe-Cr alloys, and subsequent SGPM calculations of ECI, are performed using the exact muffin-tin orbital method[29] in the coherent potential approximation. The screening parameters[21] have been calculated using supercells with quasirandom distributions of atoms. The local density approximation[30] has been used for the exchange-correlation potential. In this study, we make use of the fact that the total change of the bcc lattice parameter with composition in Fe-Cr alloys is small (less than 1%) and perform all calculations at a fixed lattice parameter of a_0=2.87 Å, which is close to the average lattice parameter for alloys containing 0–25 at. % Cr.[31]

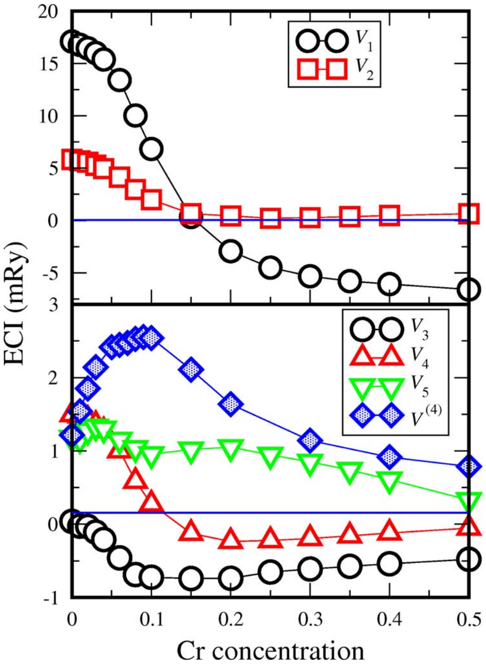

FIG. 1. (Color online) First five EPI's and the strongest four-site ECI (see text) in the FM state of Fe-rich Fe-Cr bcc alloys.

In Fig. 1, we show the first five EPI's (denoted as V_i where i is the coordination shell number) for Fe-rich Fe-Cr alloys in the FM state. It is worth noting that the EPI's are long ranged. For instance, the interaction at the 33rd coordination shell, $\mathbf{R}=a_0(\frac{5}{2}\frac{5}{2}\frac{5}{2})$, is about 0.1 mRy, which is a quite sizable value. However, the EPI's corresponding to such long distances do not affect the ASRO at the first few coordination shells and, therefore, are not considered here. The interactions included in Fig. 1 exhibit strong concentration dependencies: The first four EPI's decrease quite abruptly upon increasing the concentration of Cr, while the dependence of V_5 is more complicated.

Similarly strong and complicated concentration dependencies are exhibited by multisite ECI's, which are also found to be quite strong and long ranged in this system. For instance, in Fig. 1, we show the concentration dependence of the strongest four-site interaction, $V^{(4)}$, that corresponds to a chain of atoms aligned in the closed-packed direction: $\{(0,0,0), (\frac{1}{2}\frac{1}{2}\frac{1}{2}), (111), \text{and } (\frac{3}{2}\frac{3}{2}\frac{3}{2})\}$.[32] This strong concentration dependence also means that for the present system the widely used concentration-independent cluster expansions should be very badly convergent with respect to the order of interactions. This is not, however, the worst complication: It will be shown below that the ECI's in Fe-Cr alloys are also

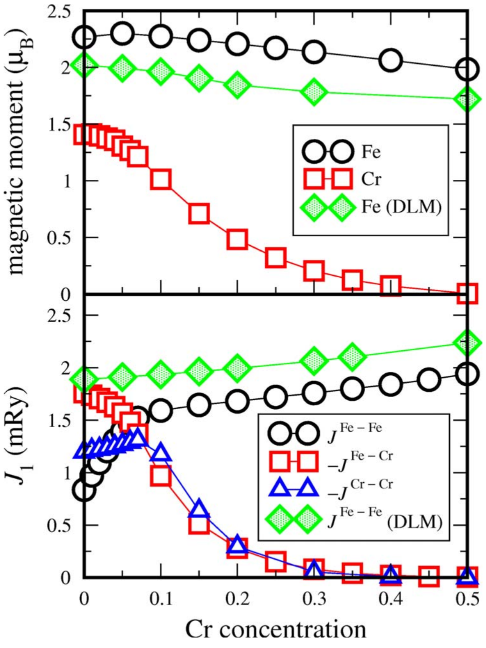

FIG. 2. (Color online) Local magnetic moment on Fe and Cr (top panel) and the NN exchange interaction parameter of the Heisenberg Hamiltonian (bottom panel) as a function of Cr concentration in Fe-Cr random alloys.

strongly dependent on the global magnetic state and, therefore, on temperature.

The strong influence of magnetism on the alloying behavior has been discussed in a number of recent papers.[15–17,19] The microscopic mechanism of this influence seems to be well understood. Thus, in the dilute limit, the Cr impurities align their magnetic moments antiparallel to those of the host Fe atoms. With the addition of Cr to the alloy, the Cr atoms that happen to be nearest neighbors have an antiferromagnetic exchange interaction with each other, causing a frustrating situation. These frustrated Fe-Cr and Cr-Cr exchange interactions lead to a gradual loss of magnetic moment by the Cr atoms with increasing Cr concentration. This is illustrated in Fig. 2 where the absolute values of local magnetic moment on Fe and Cr are plotted as a function of Cr concentration.

The figure also shows the exchange interaction parameter, J_1, for the Fe-Fe, Fe-Cr, and Cr-Cr NN pairs in random Fe-Cr alloys, calculated using the magnetic force theorem.[33] By comparing $-J_1^{\text{Fe-Cr}}$ from Fig. 2 with V_1 from Fig. 1, one notices that they have almost identical concentration dependencies. This clearly shows that the pronounced ordering anomaly in the FM dilute Fe-Cr alloys has a magnetic origin. It also follows that disorder (e.g., thermal) of the Fe magnetic moments should strongly affect the ECI by reducing the possibility for a Cr atom to align its magnetic moment antiparallel to the surrounding Fe atoms.

The completely random configuration of magnetic moments is represented in our calculations by the DLM model,[34] as a random alloy composed of spin-up (↑) and spin-down (↓) atoms.[35] In order to go continuously from the fully ordered FM state to the completely random DLM state, we employ a partial disordered local moment (PDLM) model, which formally describes a binary alloy $\text{Fe}_{1-c}\text{Cr}_c$ possessing a partial magnetic order as a three-component alloy, $(\text{Fe}\uparrow_{1-y}\text{Fe}\downarrow_y)_{1-c}\text{Cr}_c$, where $1-c$ and c are the total concentrations of Fe and Cr, respectively, and y is the relative fraction of Fe atoms having the spin-down orientation. In the PDLM state, the magnetic moments of Cr atoms are allowed to adjust their lengths and orientations to the global magnetization, which is characterized by the magnetic order parameter, $m=1-2y$.

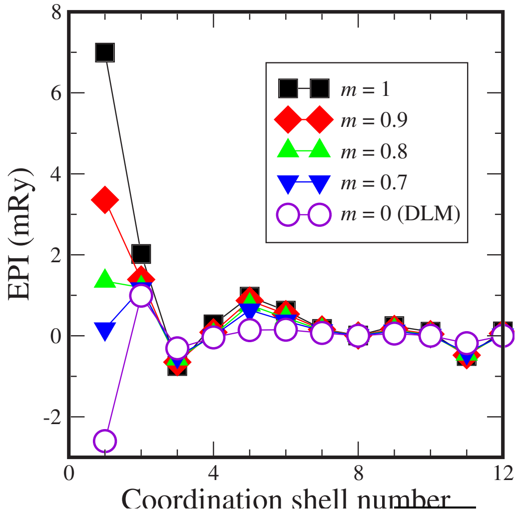

FIG. 3. (Color online) EPI in $\text{Fe}_{90}\text{Cr}_{10}$ as a function of global magnetization m.

Using the fact that thermally induced fluctuations of the local magnetic moment orientations are much faster than the atom-vacancy exchanges associated with equilibrating the atomic short-range order, one can define "spin-averaged" EPI's for a binary Fe-Cr alloy in the partial DLM state as[22]

$$\langle V_i^{\text{Fe-Cr}}\rangle = (1-y)^2\bar{V}_i^{\text{Fe}\uparrow\text{Fe}\uparrow\text{-Cr}} + 2y(1-y)\bar{V}_i^{\text{Fe}\uparrow\text{Fe}\downarrow\text{-Cr}} + y^2\bar{V}_i^{\text{Fe}\downarrow\text{Fe}\downarrow\text{-Cr}}, \tag{2}$$

where quantities such as $\bar{V}_i^{\text{Fe}\uparrow\text{Fe}\downarrow\text{-Cr}}$ are the generalized EPI's for the case of a multicomponent (ternary) alloy. They can be expressed through the ordinary EPI's (of binary combinations of the alloy components) as $\bar{V}_i^{\text{Fe}\uparrow\text{Fe}\downarrow\text{-Cr}} = \frac{1}{2}(V_i^{\text{Fe}\uparrow\text{-Cr}} + V_i^{\text{Fe}\downarrow\text{-Cr}} - V_i^{\text{Fe}\uparrow\text{-Fe}\downarrow})$.[36]

Figure 3 shows the first 12 EPI's, obtained in the SGPM calculations for $\text{Fe}_{90}\text{Cr}_{10}$ alloy, as a function of global magnetization. Although the overall change of the interactions with magnetization is moderate, the strongest EPI at the first coordination shell exhibits a substantial drop already at the onset of magnetic disorder. The origin of such strong dependence on the magnetization can be found by rewriting Eq. (2) in terms of the magnetic order parameter, using the definitions given above and $V_1^{\text{Fe}\uparrow\text{-Fe}\downarrow} = -8J_1^{\text{Fe-Fe}}$, as

$$\langle V_1^{\text{Fe-Cr}}\rangle = \frac{1+m}{2}V_1^{\text{Fe}\uparrow\text{-Cr}} + \frac{1-m}{2}V_1^{\text{Fe}\downarrow\text{-Cr}} + 2(1-m^2)J_1^{\text{Fe-Fe}}. \tag{3}$$

In the fully ordered ($m=1$) FM state, one gets $\langle V_1^{\text{Fe-Cr}}\rangle$

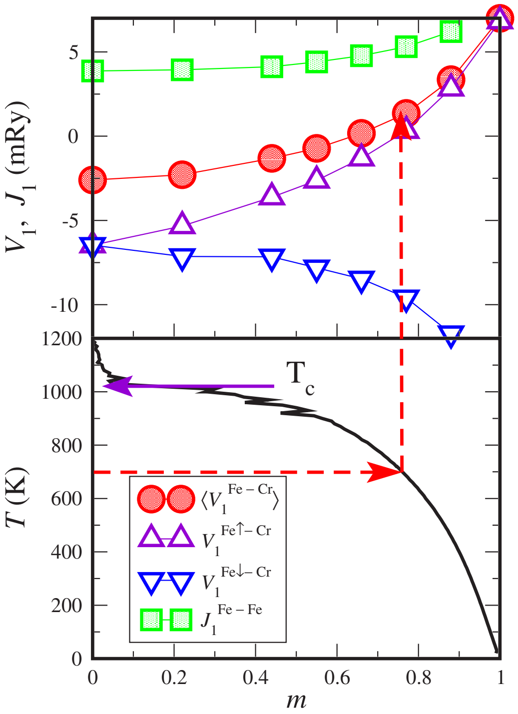

FIG. 4. (Color online) The NN EPI in $\mathrm{Fe}_{90}\mathrm{Cr}_{10}$ as a function of global magnetization m (top panel). Temperature vs global magnetization plot (bottom panel).

$=V_1^{\mathrm{Fe\uparrow\text{-}Cr}}$, whereas in the DLM state ($m=0$) the result is $\langle V_1^{\mathrm{Fe\text{-}Cr}} \rangle = V_1^{\mathrm{Fe\uparrow\text{-}Cr}} + 2J_1^{\mathrm{Fe\text{-}Fe}}$. (In the latter case, $V_i^{\mathrm{Fe\uparrow\text{-}Cr}} = V_i^{\mathrm{Fe\downarrow\text{-}Cr}}$ due to the symmetry between the spin-up and spin-down states.) This result is important in two respects. First, Eq. (3) establishes a direct connection between an EPI (which is responsible for atomic ordering) and an exchange interaction parameter (which is responsible for magnetic ordering in the system). Second, it shows that the EPI should depend on the *global* magnetic state of the system and, therefore, on the temperature.

The top panel of Fig. 4 shows the NN EPI, $\langle V_1^{\mathrm{Fe\text{-}Cr}} \rangle$, in the $\mathrm{Fe}_{90}\mathrm{Cr}_{10}$ random alloy, together with the terms from the right-hand side of Eq. (3), as a function of global magnetization. In the case of partial magnetic order, the $V_1^{\mathrm{Fe\uparrow\text{-}Cr}}$ and $V_1^{\mathrm{Fe\downarrow\text{-}Cr}}$ correspond to a NN Fe-Cr pair with the magnetic moments aligned antiparallel and parallel, respectively. These terms depend on the global magnetization through the local magnetic moment on Cr which is about $1.0\mu_B$ in the fully ordered FM state and goes to zero in the DLM state. The dependence of $\langle V_1^{\mathrm{Fe\text{-}Cr}} \rangle$ on m near $m=1$ is so strong that even little deviations from the full FM order significantly alter the NN EPI.

Let us now turn back to the ASRO measurements by Mirebeau *et al.*[13] and determine the global state of the magnetic order in the $\mathrm{Fe}_{90}\mathrm{Cr}_{10}$ alloy at the temperature, at which the ASRO developed in the samples during the annealing. In order to calculate the magnetization of an alloy under annealing, one has to perform combined Ising and Heisenberg Monte Carlo simulations because of the coupling between the atomic and magnetic order (as well as between the ECI and the J's). Such modeling would be a formidable task. Fortunately, we can use the experimental result that the ASRO in this alloy at $T=703$ K is practically zero. Therefore, we have performed Heisenberg Monte Carlo simulations for a completely random $\mathrm{Fe}_{90}\mathrm{Cr}_{10}$ alloy, also neglecting the Cr-Cr and Fe-Cr exchange interactions.

The obtained temperature vs global magnetization plot is shown in the bottom panel of Fig. 4. The calculated Curie temperature, 1020 ± 10 K, is in good agreement with the experimental value of 1020 K.[13] In the analysis below, we use the value $m=0.78$ calculated at $T=700$ K. This value is close to $m \approx 0.8$,[37] the magnetic order parameter at 700 K in pure Fe that has a similar Curie temperature of about 1040 K. Although the degree of magnetic order in the $\mathrm{Fe}_{90}\mathrm{Cr}_{10}$ alloy is still high at the annealing temperature, our calculations show that the NN EPI is reduced to about 1.0 mRy from a value of 6.9 mRy that corresponds to the same alloy at $m=1$.

This is a very small value, which is indeed very close to the border between ordering and phase separation ($V=0$), at which the specific ASRO disappears. Let us note that the absence of the ASRO was initially used just to get the magnetic state in alloy at the experimental annealing temperature of 700 K. Then, for the resulting magnetic state, we indeed have found the NN EPI, which should lead to the absence of pronounced ASRO effects. Taking into account the approximate nature of the present analysis, as discussed above, and also the fact that we have neglected, among others, the vibrational contribution to the configurational energetics (which appears to be important for the present system[38]), the calculated small value of the EPI is in very good agreement with experimental observations.[13]

Let us note that upon increasing the concentration of a random Fe-Cr alloy from 10 to 15 at. % Cr, the drop in the magnetization dependence of EPI becomes much less pronounced. This is mainly due to the decrease of the NN EPI for $m=1$ from the high value of 6.9 mRy to almost zero. The latter effect may be traced back to the reduction of magnetic moment on Cr atoms and the corresponding decrease of the Fe-Cr exchange interaction, which occur with increasing Cr concentration. The same simulation procedure as above has been used in order to obtain an estimate of the global magnetization in a random $\mathrm{Fe}_{85}\mathrm{Cr}_{15}$ alloy at 700 K. For the obtained value of $m \approx 0.75$, our GPM calculations yield $V_1 \approx -1.5$ mRy. Thus, our theory predicts a pronounced clustering tendency in the $\mathrm{Fe}_{85}\mathrm{Cr}_{15}$ alloy (the ASRO is much more sensitive to negative values of the EPI), which is indeed observed experimentally.

In summary, we have demonstrated that effective cluster interactions exhibit very strong nonlinear dependence on the global magnetization in the ferromagnetic state. Therefore, the configurational state of Fe-Cr alloys is determined by their thermal history. In particular, the experimentally observed reversion of the ordering type in the $\mathrm{Fe}_{90}\mathrm{Cr}_{10}$ alloy, as compared to the ordering type expected at absolute zero, is a consequence of the fact that in the experiment[13] this alloy was annealed at 700 K, i.e., in the incompletely ordered ferromagnetic state. In the fully ordered ferromagnetic ground state, this alloy is expected to have a quite strong atomic order.

There is probably nothing unusual in the fact that magnetism is so important for interatomic interactions in Fe-Cr alloys since both the magnetic and the chemical interactions are determined by the structure and occupancy of the *d*-electron states. More than half a century ago, Zener published his famous paper "Impact of Magnetism Upon Metallurgy."[39] For many scientists of that time, this was mostly a matter of intuition and belief. Nowadays, we have a numerical proof of the fact, that magnetism does have such an impact. It is clear that a quantitative microscopic theory of steels and other iron alloys must fully account for their magnetism. This is still a challenging task.

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