X-Ray Scattering at FeCo(001) Surfaces and the Crossover between Ordinary and Normal Transitions

Uwe Ritschel*

Fachbereich Physik, Universität GH Essen, 45117 Essen (Germany) and Fachbereich Physik, Carl-von-Ossietzky-Universität Oldenburg, 26111 Oldenburg (Germany)

Abstract

In a recent experiment by Krimmel et al. [Phys. Rev. Lett. 78, 3880 (1997)], the critical behavior of FeCo near a (001) surface was studied by x-ray scattering. Here the experimental data are reanalyzed, taking into account recent theoretical results on order-parameter profiles in the crossover regime between ordinary and normal transitions. Excellent agreement between theoretical expectations and the experimental results is found.

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*e-mail: uwe@theo-phys.uni-essen.de
Through the years, binary alloys like Fe$_3$Al or FeCo have provided one of the main test objects for theories of surface critical phenomena [1]. Some time ago, the experiment of Mailänder et al. [2,3] very impressively verified earlier theoretical predictions of Dietrich and Wagner [4] on the influence of the order parameter and the correlation function on scattering intensities near criticality. More recently, Krimmel et al. [5] studied the surface critical behavior of FeCo near a (001) surface. FeCo at or close to the ideal stoichiometry (50% Fe and 50% Co) undergoes a continuous disorder-order transition at about $T_c = 900 - 1000$ K (the critical temperature depends on the precise stoichiometry). In the high-temperature disordered phase the two species are distributed randomly on the sites of a body-centered cubic lattice (A2 phase). Below $T_c$, Fe and Co segregate on the two sublattices (B2 phase) [6]. The order parameter is proportional to the difference between the sublattice concentrations. As a consequence, the system can be modelled by an Ising antiferromagnet [7], and the corresponding bulk universality class of the transition is the one of the (ferromagnetic) Ising model. Earlier experiments on the bulk critical behavior provided good evidence for this scenario [8].

Concerning the surface critical behavior, due to missing neighbors surface spins have a reduced tendency to order such that these systems should belong to the surface universality class of the “ordinary transition” [1]. Like the previous work on Fe$_3$Al [2], the work of Krimmel et al. [3] again provided good evidence for this. The temperature dependence of the order parameter near the (001) surface, measured by surface-sensitive evanescent wave scattering [3], can be described by a power law with the exponent $\beta_1 \simeq 0.79 \pm 0.1$ [8], a value that agrees well with theoretical expectations [1,9]. However, both experiments [5,2,10] also revealed the existence of some residual long-range order near the surface for $\tau \equiv (T - T_c)/T_c > 0$, which, at the first glance, seemed incompatible with the scenario of the ordinary transition.

Theoretically the A2-B2 transition like the one in FeCo was studied in the framework of a lattice spin model [7] by Schmid [11]. The important result of Schmid was that in a system with non-ideal stoichiometry segregation effects near a (001) surface generate an ordering
(staggered) surface field $h_1$ that, even for temperatures $\tau > 0$, stabilize a residual order $m_1$ in the surface layer, in consistency with the observations of Refs. [5,2,10].

Taking into account the theoretical and experimental findings discussed above, one has to conclude that experiments of the kind reported in Ref. [5] are generically carried out in the crossover regime between “ordinary” ($h_1 = 0$) and “normal” ($h_1 = \infty$) transitions, in which the order parameter, in general, is a function $m(z)$ of the distance $z$ from the surface. At the ordinary transition, $m(z)$ vanishes identically for $\tau > 0$. For $\tau < 0$ the surface orders passively as $m_1 \sim |\tau|^{\beta_1}$ and the crossover from surface to bulk behavior, $m_{\text{bulk}} \sim |\tau|^{\beta}$, is described by a profile of the form $m(z) \sim z^{(\beta_1-\beta)/\nu}$ [12]. At the normal transition, on the other hand, the surface is completely ordered by a strong surface field. With increasing $z$, $m(z)$ monotonically decays towards the bulk value. This decay is described by the power-law $\sim z^{-\beta/\nu}$ for $a \ll z \ll \xi$ and by $\sim \exp(-z/\xi)$ for $z \gg \xi$ (where the critical exponents have their standard meaning, $a$ represents some microscopic length (lattice constant), and $\xi \sim |\tau|^{-\nu}$ is the bulk correlation length).

As discussed recently [13], any finite $h_1$ leads to a non-monotonic profile $m(z)$. The surface field provides an additional length scale $l_1 \sim h_1^{-\nu/\Delta_1}$. At bulk criticality and for $a \ll z < l_1$, $m(z)$ increases as $\sim h_1 z^{(\Delta_1-\beta)/\nu}$, before it decays to zero as $\sim z^{-\beta/\nu}$ for $z > l_1$. Slightly away from $T_c$, the scenario holds for $z < \xi$, and farther away from the surface an exponential decay sets in. For instance for $\tau > 0$ and $\xi < l_1$, the case which turns out to be relevant for the experiment, $m(z)$ increases up to $z \simeq \xi$ and then crosses over to an exponential decay [14].

Taking this crossover scenario at face value, what are the consequences for the measurable quantities in x-ray scattering experiments? A nonvanishing order parameter near the surface makes itself felt in form of an additional superstructure peak in the (001) direction of the reciprocal lattice [3,13,16]. The data of Ref. [3] for the integrated peak intensity $\hat{I}$ are displayed in Fig. 1 (symbols with error bars).

In order to calculate $\hat{I}$ from the profile $m(z)$, one may to a first approximation assume that the intensity distribution in the peak is given by [3]
\[
I(Q_z) = \left| \int_0^\infty dz \, m(z) \, e^{iQ_zz} \right|^2. \tag{1}
\]

Then for the integrated intensity the result reads

\[
\hat{I} = \int_{-\infty}^{\infty} dQ_z \, I(Q_z) = \int_0^\infty dz \left[ m(z, h_1, \tau) \right]^2. \tag{2}
\]

For the profile \(m(z)\), I choose the simple ansatz

\[
m(z, h_1, \tau) \propto \begin{cases} 
h_1 \, z^{(\Delta_1-\beta)/\nu}, & z < \xi \\
h_1 \, \xi^{(\Delta_1-\beta)/\nu} \, e^{-z/\xi+1}, & z > \xi \end{cases} \tag{3}
\]

which is sufficient for my present purpose. It yields the \(\tau\) dependence of the integral (2) for weak \(h_1\) in the regime \(l_1 \gg \xi\). This is the situation encountered in the experiment Ref. [5], because only a weak \(h_1\) is consistent with the observation of “ordinary” behavior for \(\tau < 0\) [13]. Inserting (3) in (2), the \(\tau\) dependence of \(\hat{I}\) turns out as

\[
\hat{I} \sim \tau^{2(\beta-\Delta_1)-\nu}. \tag{4}
\]

With the literature values \(\beta = 0.32, \nu = 0.63, \) and \(\Delta_1 = 0.46\) [4], this means \(\hat{I} \sim \tau^{-0.91}\), in excellent agreement with the experimental data as demonstrated by the solid line in Fig. 1.

With a similar ansatz it is straightforward to show that in the case \(\xi \gg l_1\), i.e., in a regime much closer to \(T_c\) and probably not resolved in Ref. [5], the \(\tau\) dependence of the intensity is described by \(\hat{I} \sim \tau^{2\beta-\nu}\) [17].

In contrast, Krimmel et al. [5] assumed an exponential decay

\[
m(z) \sim m_1 \exp(-z/\xi) \tag{5}
\]

of the profile and explained the vanishing of residual order above \(T_c\) with a relatively strong dependence of \(m_1\) on the temperature. The \(\tau\) dependence of \(m_1\) is described by

\[
m_1 \sim A - B \, \tau^{-\gamma_{11}}, \tag{6}
\]

with the literature value \(\gamma_{11} \simeq -0.33\) [11] and where the constants \(A\) and \(B\) were used as fit parameters in Ref. [5]. In my approach, on the other hand, the decay according to (3) is assumed to be slow and negligible for the temperature range probed in the experiment.
Just from the data it is not possible to discriminate between the two fits and, thus, the underlying theoretical models. However, there are (at least) two reasons why the scenario proposed in this work should be correct:

- The simulation results of Schmid [11] indicate a rather slow variation of \( m_1 \) as a function of \( \tau \). More precisely, the data shown e.g. in Fig. 7.c of Ref. [11] reveal that for \( \tau \approx 0.3 \), \( m_1 \) is still at about half of its value at \( \tau = 0 \). In the experiment, on the other hand, the signal vanishes for \( \tau \gtrsim 0.01 \). Hence, it is very unlikely that the decay of long-range order above \( T_c \) is attributable to the decay of \( m_1 \).

- From experiments on similar systems [18] it is known that the \( \tau \)-dependence of the measured correlation length agrees extremely well with the theoretical expectation \( \xi \sim |\tau|^{-0.63} \). With the exponential fit, where the correlation length enters as a fit parameter for the \( Q_z \) dependence of the intensity profiles, one obtains an increasing \( \xi \) upon approaching \( T_c \) (see Fig. 4.b in Ref. [5]), but with a \( \tau \)-dependence that is not consistent with the theory. In the approach proposed in this work the correct theoretical \( \tau \)-dependence enters directly in the ansatz (3) for the profile, and, as the solid line in Fig. 1 shows, the data in this case are consistent with the theory.

Finally I should like to mention that a power law similar to (4) was derived in the context of light scattering experiments by Franck [19]. In this case the reflectivity of binary mixtures near the consolute point is given the first moment of \( m(z) \) and behaves as \( \sim \tau^{\beta-2\nu-\Delta_1} \). In Ref. [19] this result was derived from scaling considerations, without explicit knowledge of the underlying order-parameter profiles.

To conclude, I have presented evidence that the results of the experiment of Krimmel et al. [5] can be explained with the crossover profiles introduced in Ref. [13]. A detailed fit of the fine structure of the superstructure reflections as presented in Fig. 3 of Ref. [4] remains to be carried out with the crossover profiles.

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Figure Caption

**Fig. 1:** The integrated scattering intensity $\hat{I}$ above the bulk critical point. The inset shows the same data in double-logarithmic form. The experimental data of Krimmel et al. are represented by the full circles. (A background intensity of 4000 was subtracted.) The fit proposed in this Communication is represented by the full line, the original fit of Ref. [1] by the dashed line.
Fig. 1