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Comment on 'Reliability of low-energy electron diffraction for studies of surface order-disorder phenomena'

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The strong multiple scattering (MS) intrinsic in LEED has led to worries[1] about its viability in probing critical properties. Thus Moritz and Lagally's recent Letter[2] (hereafter ML), calculating changes in form factors through a phase transition, is interesting, especially since it demonstrates the short-ranged character of MS. However, ML concluded that MS introduces *new* corrections to scaling, complicating the determination of critical exponents. Instead we argue that MS changes only amplitudes of corrections to scaling already present in the kinematic (single scattering) LEED intensities. How MS changes correction-to-scaling amplitudes depends on the details of correlation functions of a system and requires a painstaking calculation. The simplifying approximations of ML fail near the transition and produce artifacts. Whether the corrections to scaling cause problems in determining critical exponents is, as always, determined by how close the data are to the transition. This point is badly obscured in ML (in which the contributions of MS are never described as corrections to scaling) by the statement that the influence of MS is maximal at T_c. In fact, as with all corrections to scaling, getting closer to T_c increases the dominance of the leading singularity.

Following ML, we consider a system of adsorbed atoms on a square array of sites which has a $c(2\times2)$ phase at low T. In the kinematic approximation the intensity of the $(\frac{1}{2},\frac{1}{2})$ δ function created by the broken translational symmetry is

$$I(\tfrac{1}{2},\tfrac{1}{2}) \propto \left\langle \sum_{i\in A} n_i - \sum_{i\in B} n_i \right\rangle^2 \propto t^{2\beta}(1 + b_1 t^{\Delta_1} + b_2 t^{\Delta_2} + \ldots). \quad (1)$$

The n_i are the site-occupancy variables, and A and B refer to the two square sublattices. With MS the $(\frac{1}{2},\frac{1}{2})$ intensity becomes

$$I(\tfrac{1}{2},\tfrac{1}{2}) \propto \left\langle \left| \sum_{i\in A} f_i - \sum_{i\in B} f_i \right|^2 \right\rangle. \quad (2)$$

Here f_i is a sum of scattering amplitudes over paths involving an adatom at site i (avoiding double counting); it depends on the configuration of a finite number of adatoms surrounding site i (because of the finite electron mean free path, L). Although the $I(\frac{1}{2},\frac{1}{2})$ of Eq. (1) might be simpler to compute than that of Eq. (2), they are both legitimate definitions of the (squared) order parameter because they both vanish at T_c. Equation (2) will generally have the same correction-to-scaling exponents Δ_i as Eq. (1); only the amplitudes b_i will differ: there are no symmetry differences.

ML computes form factors for a variety of atomic configurations. These form factors are then used to predict legitimately the change in amplitude of integer beams through a transition. However ML relates the differences between Eqs. (1) and (2), $\Delta I(\frac{1}{2},\frac{1}{2})$, to the same factors. Not only is their Eq. (4) for $I(\frac{1}{2},\frac{1}{2})$ incorrect, the overall picture is also misleading. To compute $\Delta I(\frac{1}{2},\frac{1}{2})$ one needs the differences in the probabilities that certain configurations occur on A compared to their probabilities on B. The T dependence of these differences cannot be determined just from the T dependence of Eq. (1) and the overall probabilities of the configurations. Similarly, to find the changes near a transition caused by MS on the diverging diffuse intensity at the $(\frac{1}{2},\frac{1}{2})$ position, we claim that one must know correlation functions at all distances, not just the short-ranged correlations which determine changes in integer-beam intensities. Without this knowledge one cannot even estimate the changes in the b_i's caused by MS.

The phenomenological theory of phase transitions stresses the importance of length scales: the only obvious length introduced by MS is L, equal to several lattice constants. When the correlation length is much larger than L, MS will not preclude the possibility of scaling. Only if L is larger than length scales in the kinematic problem (a subtle question), does MS determine how close to T one must be to observe simple scaling. To know that one is in a scaling region, data over several decades of reduced T are needed. No LEED experiment has yet generated such data. The key limitations have been finite-size effects and limited spatial resolution,[3] not MS. We hope experimentalists will continue to probe critical properties with LEED: MS should not change the form of the function with which the data should be fitted.

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