Influence of 2s Bloch wave state excitations on quantitative HAADF STEM imaging

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In this paper the influence of 2s Bloch wave states on high angle annular dark field image contrast is studied quantitatively by Bloch wave and multislice simulations. We show that 2s states are excited beyond a critical Z value and cause a long period oscillation of the electron probe wave function that significantly influences the Z contrast. As a result, we find that the Z contrast for high Z alloys remains thickness dependent up to high sample thicknesses and this effect has to be considered for compositional analysis. In the special case of ordered alloys, the impact of 2s excitations on intensity provides a way to quantify long-range order phenomena in alloys with known composition.

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I. INTRODUCTION

Over the past decades, scanning transmission electron microscopy (STEM) has become a well-established technique for providing high-resolution structural and analytical information of solid crystalline materials. Especially the incoherent high angle annular dark field (HAADF) imaging mode, in which strong contrast related to the average atomic number of the probed material is achieved [1,2], is being used successfully for quantitative chemical composition analysis on the atomic scale. In this imaging mode, a convergent electron beam is focused on the specimen, such that the electrons propagate along a typically low order zone axis. The image is formed by electrons which are scattered into high angles and collected by an annular detector. The wave function of the propagating electron beam can be expressed in the form of eigenstates of the Schrödinger equation with the potential being formed by the 2D projection of the atomic column [3]. Because of the periodicity of the crystal potential, the columnar eigenstates can be written as Bloch waves. They are catalogued with analogy to atomic orbitals and are numbered according to a radial and angular quantum number (1s, 2p, 2s,etc.), as illustrated in Fig. 1. For a convergent STEM probe, the most relevant state that is excited is the most bound eigenstate 1s, of which the electron density amplitude is strongly peaked on the center of the atomic column. The so-called 1sstate approximation [4] has proven to give a good description of the main features of the wave function inside a crystal and the resulting STEM-HAADF image [5,6]. However, Anstis and others have shown that this approximation is not always valid and non-1s state contributions can become important for certain incident probe profiles [7] or when the column spacing is not sufficient to exclude excitation of neighboring columns [8]. Also, Rafferty et al. [9] hinted to an important contribution of the 2s Bloch wave states for high Z columns. We will show here, by means of Bloch wave calculations and multislice simulations of isolated atomic columns, that the 2s state becomes bound by the potential for atomic columns

beyond a critical mean Z value, which causes a strong deviation from the usual 1s approximation and modifies the usual description of propagation in the crystal. This has a strong impact on the resulting HAADF intensity and thus on compositional analysis, especially in ordered alloys, as it is typically performed in thicker areas of the sample. Here, the 1s channeling of the electron beam [10], which causes an oscillating variation in beam-atom interaction along the propagation direction, has died out. The averaged intensity contrast with respect to a reference material becomes then constant and the contrast scales more or less with the square of the average atomic number of the material. The validity of this approach has been proven successfully for example in the composition quantification in $Al_rGa_{1-r}N$ alloys [11,12]. In this work, however, we show that due to a substantial contribution of 2s states in high Z alloys the HAADF intensity contrast remains thickness dependent for a very large thickness range. As a result, the application of the above-mentioned method for composition analysis fails in this case. Additionally, we show that in ordered alloys the excitation of these 2s states may have a strong impact on the HAADF intensity at higher thicknesses. This provides a method to quantify the amount of long-range order in alloys of known composition on thicker samples, similar to the way the 1s channeling can be exploited to determine 3D atomic configurations on thin samples [13–16]. As ordering phenomena have been observed in different types of materials and it forms an important parameter determining material properties like mechanical, electrical, or magnetic behavior [17–19], this is a very interesting asset of the excitation of the 2s Bloch wave state.

II. METHODS

Bloch wave (BW) calculations and their spectral description are performed using our custom software B_WISE [20]. Since Bloch wave algorithms only work for plane wave illumination conditions, our software samples a number of points



FIG. 1. Right: representation of the 2D projected potential of an atomic column in a crystal, with bound (localized) and unbound (dispersive) energy eigenstates. Left: nomenclature of the columnar Bloch wave eigenstates with their spatial amplitude distribution.

within the probe: the spectra reported in this work have been calculated sampling a STEM probe obtained for a semiconvergence angle of 9 mrad and acceleration voltage of 300 kV into 2445 individual points. For each one of these points we performed a Bloch wave calculation following the original algorithm proposed by Metherell [21] and then summed up the resulting Bloch coefficients taking into account the appropriate aberration phase. Multislice (MS) simulations are performed according to [2,22] and also here a semiconvergence angle of 9 mrad and acceleration voltage of 300 kV are chosen, to match typical experimental conditions. In both the BW and MS calculations, thermal vibrations of the atoms are taken into account. For the BW calculations, this is done by considering a thermally averaged potential, while in the MS calculations the frozen phonon approximation [23] is employed. In both cases, a Debye-Waller factor of 0.2 \AA^2 is used. The frozen phonon approximation additionally naturally accounts for inelastic scattering and absorption effects [24], while these are not included in the BW calculations (no imaginary potential). Supercells for the simulations extend minimally 3×3 nm² perpendicular to the beam direction (to ensure a small enough sampling in k space) and are constructed with periodic boundary conditions in the x, y, and z directions. Therefore, when we talk about "isolated" columns, we are actually simulating a very loose lattice. The results in Fig. 2 are obtained using a sample that is formed by a simple cubic unit cell with lattice parameter a = 3 Å and a single In/Zr/Ga atom in the unit cell. For the STEM-HAADF intensity simulations, obtained by the multislice algorithm, the isolated column supercells consist of a single column with interatomic spacing of 3 Å. For the supercells of the InGaO₃ (monoclinic), In_{0.5}Ga_{0.5}N (wurtzite), and AuCu₃ (cubic) structures the interatomic spacing along the columns is 3.212 Å, 3.189 Å, and 3.753 Å and the beam direction is parallel to the [010], [1120], and [001] directions, respectively. Along the beam direction, the supercell thickness extends over approximately 30 nm and is repeated multiple times to reach the final thickness. In this way, statistical incorporation of the different constituent atoms along the columns in the case of a disordered lattice creates a sample that can be assumed to be a random configuration without producing any periodicity effects. The inner and outer acceptance angle of the HAADF detector are taken to be 35 and 270 mrad, respectively, because this corresponds to our typical experimental conditions. However, we find that our results can be generalized for different scattering angles [see Supplemental Material (SM) Figs. 1–3 [25]]. Although



FIG. 2. Left: Bloch wave excitation spectrum as a function of transverse energy (E_T) for a STEM electron probe propagating on isolated Ga (Z = 31), Zr (Z = 40), and In (Z = 49) atomic columns. Right: Multislice simulation showing a cut through the electron wave function intensity as a function of thickness. The lateral dimension of the images (y axis) is 1.5 nm and they contain five atomic columns each.

we are aware of the importance of static displacements in the analysis of alloys [12,26], we purposely remove them from our simulations to single out the effect of the 2*s* excitation on the HAADF intensity.

III. EXCITATION OF 2S STATE

To demonstrate the effect of increasing Z values on the propagating electron wave, Bloch wave and multislice simulations of a convergent STEM electron probe propagating on isolated Ga (Z = 31), Zr (Z = 40), and In (Z = 49) atomic columns are shown in Fig. 2. In the left panel, the Bloch wave excitation amplitude as a function of transverse energy (E_T) is compared for the three cases. The transverse energy [4,20,27]is defined as the difference between the z component of the electron wave kinetic energy in the sample and in the vacuum and is thus proportional to the square of the wave vector k_{τ} along the propagation direction $(E_T \sim k_z^2 - k_0^2)$. As illustrated in Fig. 1, a distinction can be made between bound (positive E_T) and unbound Bloch wave states (negative E_T). While the former are localized by the potential on the atomic columns and form sharp lines in the energy spectrum, the latter are delocalized plane waves and are contained in the excitation energy continuum at lower energies. The amount of bound states and their transverse energy depends on the depth of the potential. For the Ga column, only the 1s state is bound by the potential of the column. As the average atomic number of the column increases, the potential becomes larger and more eigenstates will start to get confined. For the Zr and especially the In column, the potential is deep enough to confine also the 2p and 2s states. We also see that the transverse energy of the states increases drastically for heavier atoms due to a stronger localization. We note that the 2s states of the Zr and heavier

TABLE I. Bloch wave properties of a STEM electron probe propagating on different types of isolated atomic columns.

	Ga	Zr	In	Ga _{0.5} In _{0.5}
$1s E_T (\text{\AA}^{-2})$	28.5	45.7	83.2	62.8
$2s E_T (\text{\AA}^{-2})$		-3.8	-1	-2.8
Unbound states (UB) $\langle E_T \rangle$ (Å ⁻²)	-8	-11	-11	-11
1 <i>s</i> +UB beating wavelength (nm)	10.8	7.2	4.3	5.5
2 <i>s</i> +UB beating wavelength (nm)		56.5	40.6	50.2

columns have negative E_T values that are very close to zero, as reported in Table I. This means that these states spread out over a very long distance and for the purpose of this paper we can safely assume them to be bound states.

The plots on the right in Fig. 2 result from multislice calculations and show a cut through the electron wave function intensity $(=|\psi|^2)$ as a function of sample thickness (on abscissas) and spatial coordinate (on ordinates) for Ga, Zr, and In columns. The electron probe is placed exactly at the center of the atomic column. Due to beating between the bound and the continuum of unbound states, oscillations in the wave function amplitude along the propagation direction arise with a frequency equal to $f = |k'_z - k''_z|$. For the continuum of unbound states, the wave vector is determined by the median of the distribution. Beating of the 1s state with the unbound states produces the short wavelength wave function oscillation at small thicknesses in each of the three cases. For Zr and In columns, the excited 2s states produce a similar beating oscillation of longer wavelength because the unbound and 2s states are closer together in energy. Apart from the amplitude oscillation centered on the column, the 2s state is also characterized by a second radially symmetric wave function amplitude maximum at a distance of approximately 0.9 Å from the column center, where the oscillation is also visible. The values reported in Table I, which are obtained from the Bloch wave calculations, show that the beating wavelength decreases as the element gets heavier, due to a stronger confinement. This agrees with the trend observed in the multislice simulations (Fig. 2, right). As described by the Bloch wave model in [20], the amplitude of the beating oscillations is damped due to a dephasing of the continuum of unbound states. The damping factor responsible for this was calculated according to [20], for the In case, and plotted as a function of thickness in the SM Fig. 4 [25]. Around a thickness of 90 nm, the unbound states start to rephase again and a second local maximum in the damping profile is reached. The same damping profile applies for 1s and 2s oscillations, and the rephasing thus causes also the reappearance of the 1s wave function oscillation with reduced intensity at higher thickness, as seen in Fig. 2. The additional effect of anomalous absorption of 1s states, however, further attenuates the 1s oscillation and therefore it dies out quicker.

The excitation of the 2p states for heavier columns is also an interesting observation. In contrast to their lack of cylindrical symmetry, they can still be excited by a cylindrically symmetric probe. Since the STEM simulations are performed by repeating a Bloch wave calculation for every incidence direction in the illumination cone, a slightly different set of Bloch states is excited for each possible incident direction and the final STEM superstate is the superposition of these single-electron states. The *p*-superstate has been calculated (illustrated in the SM Fig. 5 [25]) and turns out to have cylindrical symmetry around the atomic column on which the STEM probe is located. Most of the intensity of the *p*-type superstate is located around neighboring columns, while a node is present above the central atomic column. We can generally say that p superstates have no/neglectable intensity directly along atomic columns, neither on the central one nor on neighboring atomic columns, in any crystal system. Therefore they will give a small contribution to the HAADF intensity. However, p states will contribute to the depth dependent evolution of the total wave function/oscillations. It is clear that a threshold Z value exists for exciting 2s or 2pstates. We emphasize that this threshold depends on a variety of parameters such as the spacing between the atoms and the initial beam characteristics.

IV. IMPLICATIONS FOR QUANTITATIVE HAADF IMAGING

As we have shown that the 2s oscillation keeps the wave function amplitude strongly focused on the center of the column, we can expect it to affect scattering to the HAADF detector. Therefore, let us now move and investigate how the excitation of the bound 2s Bloch wave state is influencing the HAADF intensity as a function of sample thickness. Multislice STEM simulations are performed for two series of isolated atomic columns with increasing average atomic number to study the effect of the onset of the bound 2seigenstate. In the first series, the columns consist of one type of element with Z increasing from 22 to 49. In the second series, we start from a pure Ga column and increase the mean atomic number of the column by randomly replacing Ga by In with composition ratios ranging from 0:1. The on-column intensity output recorded by the simulated HAADF detector is plotted as a function of sample thickness in Figs. 3(a) and 3(b). For both series, the small intensity oscillations at low thicknesses (<40 nm) are caused by the 1s-unbound wave function oscillation. In a classical particle picture, this is described as the channeling effect that induces the electrons in the probe to be periodically attracted and repelled by the atomic column potential. For larger thicknesses, a strong change in the behavior of the intensity can be observed once the average atomic number along the column exceeds a certain threshold. For the low Z columns, the intensity keeps on increasing at a more or less constant rate with increasing thickness, while for the high Z columns a more steplike increase in intensity takes place. This sudden increase can be attributed to the excitation of the bound 2s Bloch wave state, for which, as illustrated before, the same threshold around $Z \approx 36$ was observed for this specific atomic spacing. The onset of this feature takes place at a thickness of around 40-60 nm, which coincides approximately with the second beating period of the 2s-unbound interaction (see Table I). This interaction causes long-wavelength intensity oscillations to persist more strongly as the average atomic number of the column increases. The fact that the same general trend is observed in both the single-atom type series and the increasing



FIG. 3. Simulated on-column STEM-HAADF intensity of isolated atomic columns for (a) columns containing a single type of element with 22 < Z < 49 and (b) columns containing a mix of Ga and In atoms in different composition ratios in a random configuration with $31 < Z_{avg} < 49$. (c) Comparison of total intensity (unit cell averaged) of a column containing an equal mix of Ga and In atoms versus the average total intensity of a pure Ga and In column.

composition series consisting of a mix of two elements indicates that the onset of confinement of the 2s state is mostly determined by the average atomic density along the column. However, just like the 1s oscillations, we expect that the 2soscillation causes some dependency of the HAADF intensity on the local distribution of atomic number density along the depth of the column. For consistency, we have compared on-column intensities and unit cell averaged intensities (SM Fig. 6 [25]) and see the same 2s attributed features in both.

Since high Z columns are strongly affected by the 2s Bloch wave state excitation, let us now see what the implications are for STEM-HAADF intensity analysis in high Z alloys. Since the STEM-HAADF intensity scales roughly with average atomic number, it is used to quantify composition of alloys by comparing the average intensity in the experimental images to that of simulated images. The composition of a layer is typically quantified in a region where the HAADF intensity ratio with respect to the reference material becomes independent of the sample thickness and knowledge of the exact thickness of the specimen is not necessary for comparison. This is nicely illustrated in Ref. [11] for $Al_xGa_{1-x}N$ alloys of different compositions. In Fig. 4, we compare similar intensity ratios in randomly configured monoclinic $(Ga_xAl_{1-x})_2O_3$ and $(In_xGa_{1-x})_2O_3$ alloys. In the case of $(Ga_xAl_{1-x})_2O_3$, the intensity ratio of (Ga_{0.5}Al_{0.5})₂O₃ and Ga₂O₃ to Al₂O₃ becomes



FIG. 4. $(Ga_xAl_{1-x})_2O_3/Al_2O_3$ and $(In_xGa_{1-x})_2O_3/Ga_2O_3$ intensity ratios for x = 0.5 and x = 1 plotted as a function of specimen thickness, with the derivatives of the curves added as subplots.

constant for thicknesses above approximately 100 nm. The strong oscillations at small thicknesses are caused by the differences in channeling behavior. However, for $(In_rGa_{1-r})_2O_3$ alloys, the desired behavior of a constant intensity ratio is not present anymore. Instead, after the channeling oscillations, the contrast doesn't saturate to a constant value, but due to the long-wavelength 2s oscillations that come into play for these heavier materials, strong contrast variations remain at large thicknesses. This effect is especially visible in the derivatives of the intensity curves, where an oscillatory behavior is clearly observed up to almost 200 nm. In the intensity ratio curves the visibility of the 2s oscillations is obscured by a second effect, which is the steady decrease of the contrast at thicknesses >100 nm. The reason for this effect is still currently under investigation. Due to this behavior, composition quantification in such heavier systems becomes a lot more difficult since specimen thickness has to be a well-known parameter to connect the intensity ratio to a composition.

V. ORDERED VS DISORDERED ALLOYS

Another important and interesting consequence of the extra jump in HAADF intensity that appears for heavy columns due to the 2s state excitation is observed in ordered alloys. Comparing ordered and disordered alloy structures, i.e., distribution of each of the constituent atoms on a distinct sublattice versus random distribution of both constituent atoms on all possible lattice sites, a significant difference in HAADF intensity occurs for large thicknesses. InGaO₃ [$(In_xGa_{1-x})_2O_3$ with x = 0.5], In_{0.5}Ga_{0.5}N, and AuCu₃ alloys are considered, to show the effect in three different lattice symmetries: monoclinic, wurtzite, and cubic (fcc), respectively. In the monoclinic lattice of InGaO₃, an equal amount of two types of lattice sites exist for the cations, which differ in their coordination to the oxygen atoms (tetrahedral vs octahedral). Due to the strong preference of the indium atoms for an octahedral environment [28], a sublattice ordering is created with all indium atoms occupying the octahedral positions and all gallium atoms the tetrahedral positions. In AuCu₃, a phase transition to an ordered state takes place below a certain transition temperature (≤ 390 °C), where the Au atoms prefer



FIG. 5. (a),(b) Schematic showing the geometry of the supercells (not full size) used for multislice STEM-HAADF simulations of ordered, consisting of pure Ga and In columns, and disordered, consisting of mixed In + Ga columns, InGaO₃ structures projected perpendicular to the beam direction (*b* axis). (c) Mean intensity as a function of thickness for an ordered vs disordered unit cell of InGaO₃, In_{0.5}Ga_{0.5}N, and AuCu₃.

to be surrounded by Cu atoms as nearest neighbor and they are positioned exclusively on the corners of the face-centered cubic (fcc) unit cell [29]. In contrast to these two intrinsic ordering phenomena, artificial ordering was assumed in the case of In_{0.5}Ga_{0.5}N. Ordered and disordered supercells with the same mean composition are constructed as described in the Methods section and the projected supercell perpendicular to the beam direction is visualized for the case of InGaO₃ in Figs. 5(a) and 5(b). The unit cell (dashed shape) and the scanning area for the simulation (filled area) are indicated. In the ordered structure, the electron beam "sees" only columns which consist of one type of atom, while in the disordered cell, each cation column consists of a random stoichiometric distribution of the two constituent atoms. This is true for each of the considered materials for the chosen beam directions. For each of the alloys, ordered and disordered, the average intensity over one unit cell (an approximate unit cell in the case of the monoclinic structure, since we can only scan rectangular cells) is extracted as a function of thickness and plotted in Fig. 5(c). The same trend is observed everywhere: for thicknesses >40 nm, HAADF intensities of ordered and disordered structures start to diverge, with the disordered lattice always having the higher intensity. The percentage difference between the ordered and disordered intensities at a thickness of 100 nm ranges between 7% and 12.5% for the three systems. The thickness onset of the divergence corresponds exactly to the characteristic thickness where the low frequency intensity oscillation caused by the excitation of the 2s state starts to dominate. To explain this remarkable phenomenon, we consider again some isolated column simulations. In Fig. 3(c), the total (unit cell averaged) HAADF intensity of a randomly

configured In_{0.5}Ga_{0.5} column is compared to that of the average of a pure Ga and In column. While in the channeling regime the intensities are still as good as equal, for larger thicknesses the randomly configured In_{0.5}Ga_{0.5} column consistently has the higher intensity. This can be understood as follows: for the pure Ga column the 2s state is not contributing, but for the mixed In_{0.5}Ga_{0.5} column and the pure In column it is (see Table I). The lack of the 2s intensity oscillation for the Ga column results in a lower intensity when averaged with an In column, compared to the mixed column. Now let us assume that, for the InGaO₃ lattice, we can approximate the total intensity as the sum of the intensities of the isolated cation columns. Since the atomic spacing along the columns in the monoclinic lattice is close to the 3 Å considered for isolated columns, the general outcome of the results can be transferred. This means that, in the ordered lattice, the 2s state is excited for only half of the cation columns (In columns), while in the disordered structure, the 2s state is excited for all cation columns ($In_{0.5}Ga_{0.5}$). Following the result of Fig. 3(c), a higher intensity should indeed be expected for the disordered lattice. The same explanation accounts for the In_{0.5}Ga_{0.5}N and AuCu₃ structures due to the mixture of one low Z (i.e., no 2sexcitation) and one high Z (i.e., 2s excitation) element. Of course, in complete lattice structures, there are more factors playing a role like cross-correlation between neighboring columns, symmetry of the lattice, etc., which will define the strength of the 2s effect. Hence every material or orientation is a very specific case and needs to be treated individually.

As a consequence of this order-disorder intensity difference, composition quantification could become complicated when the ordering in the alloy system under study is unknown.



FIG. 6. Simulated STEM-HAADF intensity at a thickness of 100 nm as a function of order parameter for $InGaO_3$, $In_{0.5}Ga_{0.5}N$, and $AuCu_3$ alloys with parabolic fit lines to the data.

However, when the composition is known, this phenomenon could possibly be exploited to our advantage. The dependency of the HAADF intensity on the ordering in the system could be used to estimate the amount of order in materials by comparing experimental STEM-HAADF images to simulations. To see if this holds up, additional supercells were created for InGaO₃, In_{0.5}Ga_{0.5}N, and AuCu₃ structures with varying degree of order. To describe the amount of order, we introduce a long-range order parameter S, as defined by Cowley et al. [30], which quantifies the number of atoms that are occupying their "correct" position in the lattice. S = 0 means a completely random distribution of atoms; S = 1 means a perfectly ordered crystal. HAADF intensities are determined at a thickness of 100 nm and their dependency to S are shown in Fig. 6. For $InGaO_3$ and $In_{0.5}Ga_{0.5}N$, a monotonic decrease in intensity is found as the order parameter increases. In the

case of AuCu₃, the intensity increases slightly as some small amount of order is introduced, but for S > 0.5 a significant and monotonic decrease of intensity is observed. A parabolic curve gives a good fit to all data sets.

VI. CONCLUSION

In conclusion, we illustrated the importance of the excitation of the bound 2s Bloch wave eigenstate in a STEM electron probe propagating on an atomic column. Just like the 1s excitation, it produces an oscillation-in this case of longer wavelength-of the electron wave function due to the interference with the unbound Bloch wave states. This oscillation is strongly reflected in the STEM HAADF intensity where it persists up to thicknesses larger than 100 nm. As a result, intensity contrast in heavy alloys is strongly modulated up to large thicknesses which hampers composition quantification. We highlighted another important consequence for alloys that consist of a mixture of elements of relatively low and high Z. When the alloy is ordered and imaged in a zone-axis direction where all columns consist of the same atom, its average intensity is systematically lower than when the alloy is disordered, starting from thicknesses ≥ 40 nm. It was shown how this dependency of the STEM-HAADF intensity on the order parameter provides a method to estimate the degree of long-range order in these types of alloy systems.

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