I. INTRODUCTION

Today, there is a strive for a controlled fabrication of nanomagnets in order to explore the concepts of spintronics at the atomic scale. Much progress was achieved in understanding direct intracenter exchange interactions in ferromagnetic few-atom clusters situated on metal surfaces (see e.g. Ref. [1] and references therein), as well as indirect surface-mediated magnetic Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions [2,3]. A central challenge remains the increasing importance of thermal fluctuations in few-atom clusters, which leads to unwanted destabilization of moments. As a consequence, in recent years the research focus has shifted towards heavy 5d transition-metal substrates, where large spin-orbit coupling (SOC) gives hope to enhance the magnetic anisotropy and to counteract superparamagnetic behavior. Indeed, for ferromagnetic Co structures on Pt(111) experiments show extraordinary large magnetic anisotropies of up to 9 meV/atom [4]. However, it has been recently realized that in transition-metal nanostructures on surfaces, SOC also induces the Dzyaloshinskii-Moriya (DM) interaction [5]. It favors non-collinear magnetic configurations and can destabilize ferromagnetism even on the atomic scale [6,7].

Less attention has been given to the lighter 4d transition metal substrates [8-14], where effects of relativistic origin such as the DM term and the magnetocrystalline anisotropy are expected to be much smaller. The exchange interaction, on the other hand, can depend critically on the hybridization with the surface and its band filling. Based on first-principles calculations it has been predicted that the nearest-neighbor (NN) exchange coupling changes from antiferromagnetic (AFM) to ferromagnetic (FM) for Fe monolayers on Ru(0001) and Rh(111), respectively [15,16]. Since its magnitude is small, interactions beyond NNs as well as higher-order terms beyond the pair-wise Heisenberg exchange, such as the four-spin and biquadratic interactions, can play a decisive role for the magnetic order [15,16]. Magnetic configurations that are surprising for Fe have been predicted for those substrates, namely a Néel state with angles of 120° between adjacent spins for Fe monolayers on Ru(0001), and a collinear double row-wise AFM udder-state on Rh(111). These two systems are thus ideal candidates to systematically study the formation of complex magnetic phases driven by frustrated interactions beyond NN Heisenberg exchange.

Here we show the essential importance of Fe 3d state itinerancy and hybridization with partially filled 4d substrate bands in monatomic-height Fe clusters of different atomic size N and various geometries. Randomly positioned single Fe atom spins in the dilute regime (N = 1) indirectly interact via the RKKY mechanism which shows inverted character on Ru(0001) and Rh(111). For Fe dimers (N = 2) we prove the AFM (Ru) to FM (Rh) cross-over of the NN exchange coupling constant J_1, and for larger clusters (2 < N ≤ 4) the onset of cluster geometry dependent compensated magnetic structures, both predicted by our first-principles calculations. We demonstrate that compact clusters are ferromagnetic while open structures exhibit compensated antiferromagnetic states. The origin of this unexpected trend arises from the competition of direct Fe-Fe exchange in the clusters and indirect exchange mediated by the substrate. Finally, we present experimental evidence for the formation of compensated spin textures both for Ru(0001) and Rh(111) in fully ordered epitaxial Fe islands.

II. EXPERIMENTAL

X-ray magnetic circular dichroism (XMCD) experiments were carried out at the ID08 beamline of the European Synchrotron Radiation Facility (ESRF), where samples can be prepared in-situ under UHV conditions.
Ru(0001) and Rh(111) single crystal surfaces were prepared by cycles of Ar-sputtering and annealing at 900°C. A scanning tunneling microscope allows to verify the cleanliness of the single crystal surfaces. Fe of 99.99% purity was deposited onto Ru(0001) and Rh(111) from a rod by electron bombardment heating. A precise calibration of the evaporator was done with the help of the scanning tunneling microscope.

During the X-ray measurements the pressure in the magnet chamber was < 3 × 10⁻¹⁰ mbar. Possible contaminations containing oxygen were excluded by monitoring the O K-edge signal at around 540 eV. X-ray absorption spectra (XAS) were measured at the Fe L₃,2-edges with 99% positive and negative circularly polarized light (σ₊ and σ₋) using the surface sensitive total electron yield (TEY) mode. XMCD and XAS signals are then defined as the difference (σ₊ − σ₋) and the average (σ₊ + σ₋)/2, respectively. The Fe L₃,2 XAS contribution to the TEY is obtained by subtraction of a background signal measured prior to Fe deposition. Spectroscopy was done at two angles of incidence with respect to the sample surface: θ = 70° (in-plane) and θ = 0° (polar). Magnetic fields up to B = 5 T are applied parallel to the X-ray beam direction. Both XAS and XMCD signals scale with the iron coverage θ. Thus, all XMCD data shown in this work are normalized to the respective L₃ peak amplitude in the non-dichroic Fe XAS. The L₃ peak value R₃ is the normalized XMCD contribution to the TEY and is shown in Fig. 1(b) and (c) for Ru(0001) and Rh(111), respectively.

III. RESULTS AND DISCUSSION

Using in-situ quench-condensed deposition of submonolayer amounts of Fe at low temperatures we achieve a statistical distribution Γ(N, g) of cluster sizes N and their respective geometries g on both Rh(111) and Ru(0001) due to suppression of diffusion of surface adatoms. Fig. 1(a) shows examples of XAS and XMCD spectra of impurities measured at B = 5 T and T = 8 K. A sharp, atomic-like dichroic signal corresponding to R₃ = 0.25 is visible, as expected for a non-saturated, thermally fluctuating single Fe atom spin moment. For comparison, saturated Fe atoms on Pt(997) give enhanced values of 0.6 under similar conditions.

The impact of an increasing Fe coverage, and thus average Fe-Fe coordination n_{Fe-Fe}, is summarized in Fig. 1(b) and (c) for Ru(0001) and Rh(111), respectively. First we note that the non-dichroic XAS L₃ peak positions shift by ∆ = 0.6 eV to higher photon energies (blue data). Positive shifts are characteristic for increasing hybridization between Fe 3d states, which leads to more efficient screening of core-hole effects during X-ray absorption. The function ∆(n_{Fe-Fe}) is usually highly non-linear, saturating already at small values n_{Fe-Fe}.

At the bottom parts of Fig. 1(b) and (c) the evolution of the magnetic signal with raising n_{Fe-Fe} is shown. The trend of R₃ θ for Fe on Ru(0001) shows a steady decay of the average magnetization with Fe coverage, indicating progressive magnetic compensation. Comparing the R₃ values for θ = 70° and θ = 0°, we observe an in-plane magnetic anisotropy in the range θ < 0.1 ML. For Fe clusters on Rh(111) we find an even steeper initial decrease of R₃ with coverage and at low coverages an in-plane magnetic anisotropy. In contrast to Ru(0001),...
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method as implemented in the Vienna Ab-Initio Simula-
surfaces. We have applied the projector-augmented-wave
(DFT) for Fe clusters of different size and shape on both
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two Fe adatoms as a function of their distance. The ex-
fore, we first focus on the exchange interaction between
exchange interaction mediated by the substrate. There-
symbols mark pure fcc sites and mixed dimers. The inset
shows the position of the Fe atoms in the dimer. The fitting
has been performed with an RKKY-like function.

at an intermediate coverage $\theta_m \sim 0.25$ ML the magne-
tization reaches a minimum and increases monotonously
thereafter.

In order to understand the observed trends of the
magnetization with coverage, we have performed first-
principles calculations based on density functional theory
(DFT) for Fe clusters of different size and shape on both
surfaces. We have applied the projector-augmented-wave
method as implemented in the Vienna Ab-Initio Simula-
tion Package (VASP) and used the generalized gra-
dient approximation (GGA) to the exchange-correlation
functional. We consider different collinear magnetic
configurations of the clusters and compare their total en-
ergies taking vertical and lateral structural relaxations
into account. Computational details can be found in the
Appendix A.

At low coverage there will be mostly a distribution of
single adatoms which can interact with each other via the
exchange interaction mediated by the substrate. There-
fore, we first focus on the exchange interaction between
two Fe adatoms as a function of their distance. The ex-
change constants $J(r)$ obtained from total energy calcu-
lations are presented in Fig. 2. As expected we observe
an oscillatory behavior of $J(r)$ changing from FM ($J > 0$)
to AFM ($J < 0$) and a decay of its magnitude with in-
creasing Fe-Fe separation. Interestingly, the trend found
for Fe dimers on the Rh and Ru surface is almost per-
factly inverted. In contrast to the exchange interaction
reported for substrates with a filled $d$-band we find
that the NN exchange constant $J_1$ is reduced by about
one order of magnitude and, thus, in competition with
indirect exchange interactions $J_n$ with $n > 1$. The latter
will in the following be referred to as $J_{RKKY}$.

Our calculations for Fe trimers and tetramers on
Rh(111) and Ru(0001), shown in Fig. 3(a) and (b), re-
spectively, display a complex dependence of the mag-
netic order on the cluster geometry. For Rh(111) we find
that compact trimers and tetramers possess a FM ground
state which is in accordance with the FM NN exchange
coupling from the dimer calculations (cf. Fig. 2) although
the energy differences are much larger than expected
from the exchange constants obtained from the dimers.
However, Fe clusters in an open structure show a ten-
dency to AFM order with compensated spin structures.
This is surprising in view of the FM exchange interaction
of the dimers. Interestingly, the open tetramers already
display the $uudd$ state predicted for the full monolayer.

These effects arise due to a competition of direct Fe-Fe
exchange and indirect exchange mediated by the sub-
strate which are closely linked with the cluster geom-
etry and structural relaxations that differ for open and
compact structures. The impact of the structural rel-
xation on the magnetic state is evident from Fig. 3(a)
if one compares the energy differences obtained without
taking structural relaxations into account. In the case of
a NN dimer on Rh(111) the exchange energy is reduced
by one order of magnitude upon relaxation, leading to the
very low value of 6 meV/Fe-atom. A considerable reduc-
tion of the energy difference occurs also for the compact
trimers and tetramers. For most of the open cluster con-
figurations, the energetically favorable state even changes
from a FM to a compensated state upon taking structural
relaxations into account.

A similar trend of magnetic order is found for Fe
trimers and tetramers on Ru(0001) as shown in Fig. 3(b).
For open structures, compensated AFM spin structures
are found which is expected from the AFM NN exchange
in Fe dimers (cf. Fig. 2). Note, that the AFM NN ex-
change is driven by the hybridization with the substrate.
This can be seen by comparing the energy differences for
the Fe dimer without structural relaxation which prefers
a FM state (see Fig. 3(b)). However, compact trimers
and tetramers are in a FM ground state. The origin of
this unexpected change of exchange coupling in the clus-
ters is due to the enhanced direct FM Fe-Fe exchange
interaction and a weakened effect of the Ru substrate.

In order to obtain a quantitative interpretation of
our experimental data based on the magnetic config-
urations calculated from first-principles we performed
Monte-Carlo (MC) simulations. Knowing the magnetic
ground states of all cluster configurations $(N, g)$ with
$N \leq 4$, MC simulations of $\Gamma(N, g)$ allows us to esti-
mate the coverage dependent average magnetization of
the ensemble in a magnetic field $B = 5T$. We assume
each cluster to be magnetically independent and that
each single adatom interacts only with one closest sin-
gle atom via the RKKY interaction as described in the
supplementary. The magnetic contribution of a certain
cluster with $(N, g)$ to the total signal $R_\text{L3}$ is then given
by a Boltzmann statistics weighted according to $\Gamma(N, g)$,
where also induced substrate moments enter the Zeeman
energy term (see Appendix B for further details).

The low coverage behavior ($\theta < 0.1$ML) of $R_\text{L3}$ shown
in Fig. 4 can be understood based on the RKKY interac-
tions and the NN exchange constant $J_1$. In the simpler

![Graph](attachment:image.png)

**FIG. 2:** Calculated exchange coupling constants $J(r)$ as a
function of separation $r$ between pairs of Fe adatoms. Open
symbols denote results for pure hcp adsorption sites and filled
symbols mark pure fcc sites and mixed dimers. The inset
shows the position of the Fe atoms in the dimer.
FIG. 3: Total energy differences between different magnetic configurations for the Fe dimer, trimers and tetramers on (a) Rh(111) and (b) Ru(0001). Energy differences in meV per Fe atom are given with respect to the FM state. Values in brackets are energy differences without taking structural relaxations into account.

case of Ru(0001) the magnetization trend at low coverages is dominated by the AFM NN exchange constant $J_1 < 0$. In Fig. 3(b) the result for magnetically independent clusters excluding RKKY interactions is shown, which reproduces the continuous decay of the magnetization well, considering that the modeling contains no free parameter. At lowest coverage, $R_{L3}$ corresponds to a single spin moment of 3.0$\mu_B$ as obtained from our DFT calculations in the corresponding Zeeman field.

Turning to the case of Fe clusters on Rh(111), we find that $R_{L3}$ at lowest coverages is larger compared to the values found for Ru(0001), which we attribute to (i) the enhanced spin moment 3.2$\mu_B$ of a single Fe atom on Rh(111) and (ii) the about ten times larger magnetic susceptibility of Rh(111) leading to larger induced substrate moments. The latter enter the Boltzmann statistics via the Zeeman term and stabilize the Fe spin moments. It is evident that even a qualitative understanding of the trend $R_{L3}(\theta)$ based on the NN exchange interaction is impossible in the case of Rh. The steep decrease of $R_{L3}$ at lowest coverages is surprising in view of the positive NN exchange coupling $J_1$. Starting from single atoms the increase of $\theta$ should thus enhance the average magnetization per Fe atom due to FM dimer formation as seen by the dashed curve in Fig. 3(c). However, if we take into account the RKKY coupling between single Fe atoms on Rh(111) we observe that the AFM exchange coupling for separations of up to 6 Å overcompensate by far the contribution of the FM NN dimer coupling and accurately reproduces the steep decrease of the average magnetization below $\theta = 0.1$ML (solid curve in Fig. 3(c)).

At intermediate coverages, mostly the formation of FM dimers and FM compact trimers on Rh(111) leads to a plateau in $R_{L3}$ in good agreement with our experimental data. According to statistics, trimer configurations start to play a role at coverages $\theta > 0.1$ML, which again suppress the average moment due to intrinsic compensated structures (see Fig. 3). For the excellent quantitative agreement between experiment and simulation, the geometry dependent ground states of tetramers as obtained from DFT are nevertheless important. This is visible in the two simulations shown in Fig. 3(c) for comparison in which it has been assumed that all trimers and tetramers are either perfectly FM or in a maximum compensated magnetic state.

Our simulations are valid up to coverages of about 0.3 ML. Beyond that the simulated values $R_{L3}$ start to decrease due to the increasing spectral weight of clusters with $N > 4$, which in our MC simulations are assumed to have zero moment (see Appendix B). We attribute the rise of the experimental $R_{L3}$ signal to the formation of three-dimensional FM clusters which are less coupled to the substrate and thus will be dominated by the FM direct exchange between Fe moments.

Finally, we present experimental evidence for compensated magnetic ground states of Fe MLs on the hexagonal surfaces Ru(0001) and Rh(111). From the data discussed
so far only the measurements on Ru(0001) are compatible with such a compensated ground state, since the values for $R_{L_3}$ reach very low values at high coverages $\theta = 0.5\text{ML}$ (see Fig. IIb). For Rh(111) it is evident that such a state cannot be reached by quench-condensed deposition. This is not really surprising since the structure is expected to be disordered, and beyond NN corrections are strongly hampered in a random fashion. We therefore test our systems in the presence of structural order.

In Fig. II (a)-(b) STM topographies of 5-10nm wide monatomic-height Fe islands on Ru(0001) and Rh(111) are shown, respectively, which grow epitaxially at deposition temperatures of $T = 300\text{K}$. On Ru(0001) triangular shaped islands with 5-10nm in diameter are formed on the terraces, and smaller islands decorate the terrace step edges. The onset of the 2nd layer formation on the islands is only visible on Ru(0001), but the ratio between bilayer and monolayer areas corresponds to less than 5%. On Rh(111), islands of mostly truncated triangular shape are randomly distributed.

Fig. II(c) shows $R_{L_3}$ for the two systems measured at $B = 5\text{T}$ and different temperatures. At $T = 8\text{K}$ only a small Fe dichroic signal of $R_{L_3} = 0.07$ and $R_{L_3} = 0.09$ is present for $\theta = 0.1\text{ML}$ on Ru(0001) and Rh(111), indicative of intrinsically compensated magnetic ground states in both cases. The temperature dependence up to $T = 300\text{K}$ can be fitted by a classical Boltzmann statistics of a constant superparamagnetic macrospin $M_{Fe}$, suggesting stable ground states up to energy scales beyond 25meV. As in the quench-condensed samples a faint in-plane magnetic easy direction is observed for both substrates (see Fig. II b) and (c), where open/full triangles correspond to $\vartheta = 0^\circ$ and $\vartheta = 70^\circ$).

The difference of the island results compared to those obtained by quench-condensed deposition underlines the importance of the structure on the magnetic state. The stabilization of a compensated magnetic configuration on Rh(111) against a FM exchange term $J_1 > 0$ is only possible for ordered compact clusters, which allow effective hybridization of Fe 3d states over larger distances. Increased hybridization in the ordered case is also directly visible in the measured XAS $L_3$ peak photon energy, which remains $\sim 0.4\text{eV}$ above the value of quench-condensed structures at largest coverages (see Fig. II b) and (c), top).

IV. CONCLUSIONS

In conclusion, we have shown a complex trend of magnetic order in Fe nanostructures on 4d transition-metal surfaces due to the hybridization of Fe 3d-states with the partly filled substrate 4d-band. For Fe dimers the nearest-neighbor exchange is very small and of opposite sign on the Ru and Rh surface. For larger clusters the competition of direct FM Fe-Fe exchange with the indirect exchange mediated by the substrate determines the magnetic order. Finally, we have presented first experimental evidence for the formation of compensated spin textures in epitaxial Fe islands on both for Ru(0001) and Rh(111) as predicted by theory.

Acknowledgments

F. O., P. F., and S. H. acknowledge financial support by the Deutsche Forschungsgemeinschaft within the SFB 677 and thank the HLRN for providing high-performance computing resources. J. H. acknowledges the Czech Purkyně fellowship program.

Appendix A: Computational details

Fe clusters on Rh(111) and Ru(0001) have been studied based on density functional theory calculations in the generalized gradient approximation (GGA) to the exchange-correlation functional, using the projector-augmented-wave method as implemented in the Vienna Ab-Initio Simulation Package (VASP). All calculations have been performed in the scalar-relativistic approximation, i.e. neglecting the effect of spin-orbit coupling. To model the Fe clusters we have used the $p(4 \times 4)$ and $p(5 \times 5)$ surface unit cells for clusters with $N < 4$ and $N = 4$, respectively. To model the Rh(111) or Ru(0001) surface eight layers have been used. The adatoms as well as the two upmost surface layers have been structurally relaxed until the forces were smaller than 0.005 eV/Å. A (5 × 5 × 1) and (3 × 3 × 1) Γ-centered k-point mesh has been used for the $p(4 \times 4)$ and $p(5 \times 5)$ surface unit cell, respectively. The experimental lattice constant of 3.8034 Å for Rh and lattice parameters of 2.7059 Å and 4.2815 Å for Ru have been chosen. The energy cutoff parameter for the plane wave expansion was 390 eV and a Gaussian smearing of $\sigma = 0.07$ eV has been applied.

An important aspect of our approach to calculate the exchange constants is the possible interaction of the clusters with those in adjacent cells due to the two-dimensional (2D) periodic boundary conditions. In order to estimate the influence of atoms in neighboring cells we have performed test calculations in the $p(3 \times 3)$, $p(4 \times 4)$, and $p(5 \times 5)$ unit cells. We found that the $p(4 \times 4)$ unit cell size is sufficiently large to avoid spurious interaction effects for compact clusters and to determine their magnetic ground state. However, if the distance between the adatoms within the unit cell is large as for dimers with large separation, the influence of atoms in the adjacent unit cells also becomes important. We have taken such interactions into account when determining the RKKY exchange constants. More detailed information can be found in Ref. [32].
Appendix B: Monte Carlo Simulations

During the Monte Carlo (MC) simulation iron atoms are randomly deposited onto two hexagonal hcp and fcc sublattices, each of which have a size of 500 × 500. Since in the experiment the atoms are deposited at a temperature of \( T = 8 \) K no thermal activated hopping of atoms is included in the simulation. However, we do take into account random tip-over processes onto neighboring free adsorption sites, if the initial MC step chooses a landing site which is already occupied by an iron atom. During one MC deposition cycle the sum of the number of atoms on both sublattices is increased by 0.02\% of a full monolayer (ML).

After every MC deposition cycle the program counts the different types of clusters: an atom is evaluated as a monomer if it has no nearest neighbor (NN) on the same lattice, two atoms are evaluated as a dimer if they have just themselves as NNs, and so on. Moreover, the program distinguishes between different geometries \( g \) for one and the same cluster size \( N \), e.g. between linear trimers and trimers with an angle. The MC simulation thus gives access to the distribution \( \Gamma(N,g) \) of all different cluster configurations \( (N,g) \) from monomers to tetramers \( (N < 5) \) both on hcp or fcc sublattices. Figure 5(a) reflects the statistics of cluster counts with size \( N \) versus coverage.

Our DFT calculations show the importance of long-range RKKY interactions between pairs of monomers (see Fig. 2), which become important in the lowest coverage range. To capture these effects the statistics of single atom pairs is extracted, evaluating the combination of monomer pairs on hcp and fcc lattices from 2nd NN up to 5th NN distances for a given MC distribution. Herby, we only count pairs for which other monomers are found only at larger distances. This approximation thus assumes that for these pairs, residual oscillating RKKY field contributions of other surrounding monomers and pairs in average cancel each other and play a minor role.

Figure 5 illustrates the evolution of \((N,g)\) with coverage. Figure 5(a) shows the statistics of cluster counts with size \( N \) versus coverage, while Fig. 5(b) translates this statistics into spectral weights contributing to the X-ray absorption signal. The spectral weight \( \omega(N) \) is hereby defined as:

\[
\omega(N) = \frac{\sum_{N,g}^g N \cdot \Gamma(N,g)}{\sum_{N,g}^g N \cdot \Gamma(N,g)}
\]

The degree of magnetic alignment of a given Fe cluster \((N,g)\) (including RKKY-coupled pairs of monomers) with an applied field of \( B = B \hat{z} \) is estimated using a Zeeman energy term of the form \( E(M_{tot}^{N,g},B,\Theta) = -B \cdot M_{tot}^{N,g} \cdot \cos(\Theta) \), where the absolute value of the total moment vector \( M_{tot}^{N,g} \) in units \( \mu_B \) is defined as the sum of total Fe moment \( M_{Fe} \) and induced substrate moments \( M_{tot}^{N,g} = M_{Fe} + M_{4d} \). \( \Theta \) is the angle between the moment vector and the field direction \( \hat{z} \). All moments are readily taken from DFT results. The contribution \( R_{L_3}^{N,g}(B,T) \) of a certain cluster with \((N,g)\) to the total signal \( R_{L_3} \) is then given by a Boltzmann statistics, allowing \( M_{tot}^{N,g} \) to point in all directions in space:

\[
R_{L_3}^{N,g}(B,T) = \frac{R_{L_3}^{tot}}{N \cdot 3 \mu_B} \int_0^\pi \int_0^\pi M_{Fe}^{N,g} \cdot \cos(\Theta) \cdot \sin(\Theta) \cdot e^{-E(M_{tot}^{N,g},B,\Theta)/k_B T} d\Theta d\varphi / Z
\]

where \( Z \) is the partition function. The term \( M_{Fe}^{N,g} \cdot \cos(\Theta) \) projects the Fe cluster moments \( M_{Fe}^{N,g} \) onto the \( \hat{z} \) direction, which accounts for the fact that the XMCD technique measures Fe moment components along the X-ray beam direction. The calibration factor in front of the integral contains \( R_{L_3}^{tot} = (0.6 \pm 0.05) \), which is the value of \( R_{L_3} \) expected for low coordinated Fe spin moments \((3.0 \pm 0.2) \mu_B \) in a saturating magnetic field\[1\]. In our simulation we thus make the assumption that the value \( R_{L_3}^{tot} \) is a Fe moment dependent constant value which does not change significantly if the coordination state of the Fe changes. This assumption is not generally valid but is a good approximation for Fe atoms in metallic environments. For a comparison with the experimental spectroscopy data, the total simulated signal \( R_{L_3} \) is defined as the sum of all components \( R_{L_3}^{N,g}(B,T) \) of clusters \((N,g)\) at experimental conditions \( T = 8 \) K and \( B = 5 \) T, scaled to their respective spectral weights determined by \( \Gamma(N,g) \).

The coverage dependent spectral weight of clusters \( N = 1, 2, 3, 4 \) given in Fig. 5(b) shows that the coverage range up to \( \theta = 0.1 \) ML is clearly dominated by monomers and dimers as expected (grey shaded region). At \( \theta = 0.2 \) ML dimer contributions with \( N = 2 \) are comparable to those of monomers. At the same time also clusters with \( N = 3 \) gain a spectral weight of more than 10\%, indicating the onset of larger cluster contributions.

At coverages \( \theta = 0.25 \) ML the spectral weight of all cluster contributions with \( N > 4 \) reaches a value of 10\%. In the framework of our simulations these contributions
FIG. 5: (a) Distribution $\sum_g \Gamma(N,g)$ of cluster sizes $N$ versus coverage and (b) spectral weights $\omega(N)$ of the different cluster sizes $N$.

are considered to have zero average moment. As a consequence the simulation represents a lower limit of the expected signal especially for larger coverages. From the statistics we estimate the validity of our MC simulation to be limited to the coverage range $\theta < 0.3$ ML, also because beyond this coverage we expect the onset of intermediate and 2nd layer formation, both not covered in our MC simulation.

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