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## Core-Level Photoemission and Work-Function Investigation of Na on Cu(110)

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## Core-level photoemission and work-function investigation of Na on Cu(110)

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Core-level photoemission, low-energy electron diffraction (LEED), and work-function change measurements have been carried out to study the coverage dependence of Na/Cu(110) at room temperature. The results of LEED and work-function measurements are qualitatively similar to most other investigations of alkali-metal adsorption on fcc(110) metal surfaces. With LEED, we observed an alkali-metal-induced  $(1 \times 2)$  reconstruction at intermediate coverage. We have performed a simple calculation to account for the work-function differences between Na/fcc(110) and Na/fcc(111) metal surfaces. The comparison of coverage-dependent core-level binding-energy shifts between Na/Cu(110) and Na/Cu(111) reveals that a low-coverage plateau in the curve of binding energy vs Na coverage for Na/Cu(110) is associated with the Na-induced reconstruction, and can be accounted for within a localized picture of the reconstruction.

### I. INTRODUCTION

The  $(1 \times 2)$  alkali-metal-induced reconstruction of (110) fcc metal surfaces has been extensively investigated both experimentally and theoretically over the past few years.<sup>1-7</sup> A missing row structure is now generally accepted as the most favored model of the surface reconstruction based on low-energy electron diffraction (LEED) intensity analysis,<sup>8,9</sup> ion scattering,<sup>10,11</sup> and scanning tunneling microscopy<sup>12</sup> for a number of alkali-metal/fcc-metal (110) systems, such as Li/Cu(110),<sup>10</sup> K/Cu(110),<sup>12,13</sup> Cs/Cu(110),<sup>13</sup> K/Ag(110),<sup>11</sup> K/Pb(110), and Cs/Pb(110).<sup>8,9</sup> However, the reconstruction mechanism, its driving force, and whether the reconstruction is a result of local or long-range forces remain under discussion. In this paper we would like to present the results of an investigation of the system Na/Cu(110) using the techniques of LEED, work-function measurements, and core-level photoemission spectroscopy. Core-level photoemission studies make it possible to determine the number and nature of different bonding sites, and may provide an indirect test of existing reconstruction models. In addition, we can make a valuable comparison to our previous photoemission results of Na/Cu(111).

### II. EXPERIMENT

The experiments were performed either at the University of Rhode Island (URI) or at beamline U14A of the National Synchrotron Light Source, Brookhaven National Laboratory (BHL). The base pressure of the ultrahigh-vacuum systems in both cases was in the low- $10^{-10}$  Torr range. The Cu(110) sample was cleaned by a combination of Ar<sup>+</sup> sputtering and annealing at 450 °C. Surface cleanliness was monitored by Auger electron spectroscopy, LEED, or core-level photoemission. The Na was deposited from a thoroughly outgassed commer-

cial SAES getter source equipped with a shutter and collimation, with pressure rises during evaporations of  $< 5 \times 10^{-10}$  Torr. All evaporations and measurements were performed at room temperature.

The photoemission data to be presented here were obtained with a photon energy of 70 eV. The photoelectrons were detected by an angle-integrating double-pass cylindrical mirror analyzer (CMA) using a pass energy of 25 eV. The overall energy resolution was  $\sim 0.3$  eV. Work-function measurements were performed either using the retarding field method with a LEED system at URI or the secondary cutoff of photoelectrons at BHL.

### III. RESULTS

#### A. Work-function and LEED measurements

Figure 1 shows the work function for Na/Cu(110) as a function of Na coverage along with corresponding LEED patterns. The coverage of one monolayer (ML) was determined by the appearance of a second layer peak in the Na 2*p* core-level spectra. We could not determine the absolute density of Na at saturation because no ordered structure associated with the Na overlayer was observed. As shown in Fig. 1, the work function decreases linearly with Na coverage from the clean-surface value of 4.48 eV, reaches a minimum of 2.33 eV at  $\theta = 0.55$  ML, and then increases to saturation at 2.58 eV at the completion of the first layer. This general work-function behavior is consistent with that of almost all systems of alkali metals/metals. Comparing it to our previous work-function results of Na/Cu(111) (Ref. 14) (shown by the solid line in Fig. 1), we find that the work-function curve of Na/Cu(111) has a deeper minimum than for Na/Cu(110). This is similar to the results of Na/Ni(111) versus Na/Ni(110), as reported by Gerlach and Rhodin<sup>15</sup> (see Table I).

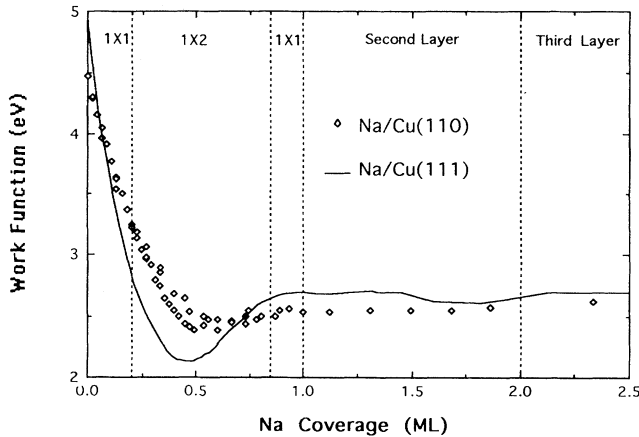


FIG. 1. Work-function change of Na/Cu(110) as a function of Na coverage along with corresponding LEED patterns. The work function of Na/Cu(111) is included for comparison as a solid line in the figure and is taken from Ref. 14. Note the offset at zero coverage due to the different absolute work functions of clean Cu(110) and Cu(111).

The clean Cu(110) surface at room temperature exhibits an unreconstructed ( $1 \times 1$ ) LEED pattern. At Na coverages below 0.2 ML, the LEED pattern remains ( $1 \times 1$ ). With increasing coverage, elliptical half-order spots in the [001] direction, with the long axis along the [001] direction, gradually appear. This elliptical ( $1 \times 2$ ) diffraction pattern may be related to the ( $1 \times 3$ ) superstructure observed by Fan and Ignatiev<sup>17</sup> in their studies of K/Cu(110) and Cs/Cu(110). In our study, a sharp ( $1 \times 2$ ) LEED pattern is first observed for a coverage of about 0.3 ML. If the coverage is further increased, the LEED pattern becomes ( $1 \times 2$ ) with additional streaks along [001] near the  $(\pm \frac{1}{2}, 0)$  positions. After the coverage reaches 0.75 ML, the half-order spots and the additional streaks become weaker. Only a ( $1 \times 1$ ) LEED pattern is observed for coverages greater than 0.85 ML. Similar sequences of LEED pattern changes with increasing coverage have also been observed for K, Cs/Cu(110) (Ref. 17) and Na, K, Cs/Ni(110).<sup>18</sup>

#### B. Core-level photoemission measurements

In Fig. 2 a sequence of spectra of the Na  $2p$  core level for Na/Cu(110) is plotted as a function of Na coverage. At an Na coverage of 0.04 ML, we observed a peak at a binding energy of 31.05 eV. With increasing Na cover-

age, this peak shifts towards lower binding energy. The peak position drops to a binding energy of 30.54 eV when the Na coverage reaches 1 ML. For Na coverages above the first layer, a second Na peak emerges on the higher-binding-energy side of the first layer peak. As for our previous measurements of Na/Cu(111),<sup>14</sup> we observed no evidence of intermixing between Na and the substrate, which had been reported for the systems Na/Al(111) and Na/Al(100) by Andersen *et al.*<sup>19</sup>

The coverage dependence of the binding energy of the Na  $2p$  core level for Na/Cu(110) is presented in Fig. 3, along with results of our previous investigation of Na/Cu(111) for comparison (Fig. 4). For Na/Cu(110), in contrast to Na/Cu(111), a definite plateau in this curve is observed at low Na coverages. After the Na coverage reaches 0.2 ML, the binding energy drops continuously up to 1 ML for Na/Cu(110). The total-binding-energy shift is 0.56 eV from  $\sim 0.02$  ML, the smallest coverage we investigated, to 1 ML for Na/Cu(110), which is smaller than the overall shift of 0.93 eV between  $\sim 0.02$  and 1 ML for Na/Cu(111). Note that the binding energy of the core level in the second layer is independent of substrate face.

#### IV. DISCUSSION

The results of a recent scanning tunneling microscopy (STM) study of K/Cu(110) by Schuster *et al.*<sup>12</sup> demonstrate the local nature of the  $K$ -induced reconstruction of Cu(110). According to their results, at small  $K$  coverages, dispersed stable nuclei in which a single  $K$  adatom replaces two to three adjacent Cu atoms along  $[1\bar{1}0]$  are formed on the flat terraces. Increasing the coverage leads to preferential growth of nuclei in the  $[1\bar{1}0]$  direction; at the same time the average separation of the troughs in the [001] direction decreases. With a further increase in the  $K$  coverage, the missing-row reconstruction goes to completion. We would like to make use of these observations to explain our work-function and core-level binding-energy results. It is also worth pointing out the similarity between the embedded nature of the alkali metal in the missing-row (MR) reconstructed fcc(110) systems such as Na/Cu(110) to the substitutional adsorption of alkali metals on aluminum surfaces.<sup>19,20</sup>

#### A. Work function

The work-function decrease induced by alkali-metal adsorption on a surface can be ascribed to the formation

TABLE I. Work-function data of Na on copper and nickel surfaces.

	Na/Cu(110)	Na/Cu(111) <sup>a</sup>	Na/Ni(110) <sup>b</sup>	Na/Ni(111) <sup>b</sup>
$\Phi_{\text{clean}}$ (eV) <sup>c</sup>	4.48	4.94	5.04	5.35
$\Delta\Phi_{\text{min}}$ (eV) (expt.)	-2.15	-2.75	-2.1	-3.05
$\Delta\Phi_{1\text{ML}}$ (eV) (expt.)	-1.9	-2.25	-1.95	-2.25
$D_0$ (Db)	2.66	4.26	3.2	7.4

<sup>a</sup>Reference 14.

<sup>b</sup>Reference 15.

<sup>c</sup>Reference 16.  $\Phi_{\text{clean}}$  is the absolute work function of the clean substrate.

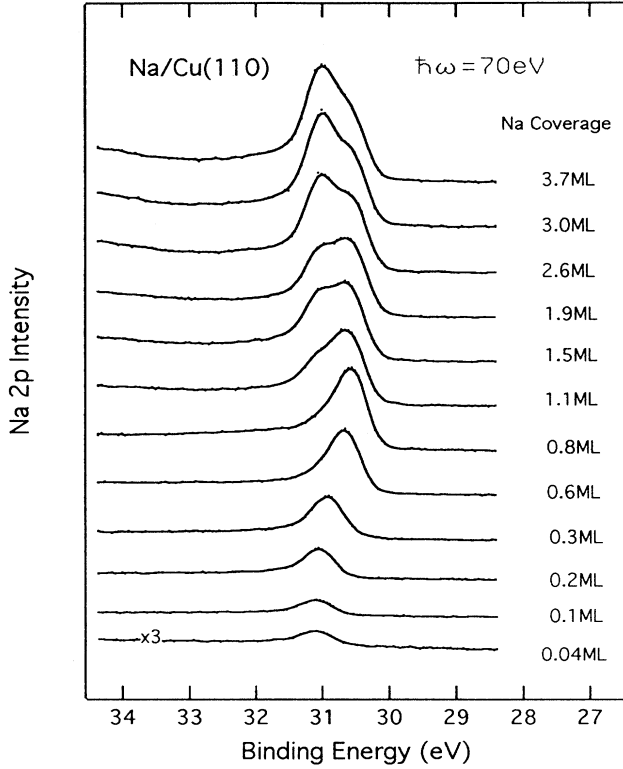


FIG. 2. Na  $2p$  core-level photoemission spectra for varying Na coverages on Cu(110).

of an electric dipole layer at the surface which is antiparallel to that of the substrate surface dipole layer. With increasing coverage, mutual depolarization of the dipoles of the alkali-metal overlayer leads to a continuous decrease (in some models) of the dipole moment per particle, and to the occurrence of a minimum in the work-function-change curve.<sup>21</sup>

The work-function change  $\Delta\Phi$ , due to alkali-metal adsorption, can be written in the following general form:<sup>22,23</sup>

$$\Delta\Phi = D(\theta) \times \theta \times \sigma_{\text{ML}} / \epsilon_0, \quad (1)$$

where  $D(\theta)$  is the (coverage-dependent) dipole moment

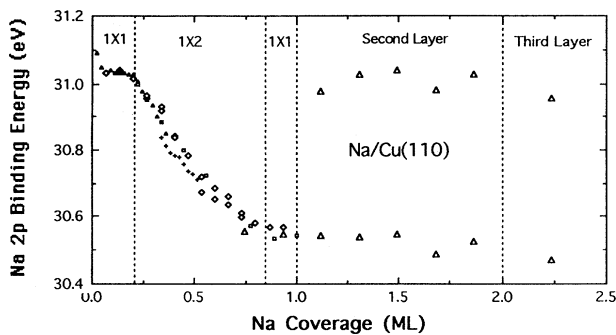


FIG. 3. Coverage dependence of the Na  $2p$  core-level binding energy for Na/Cu(110).

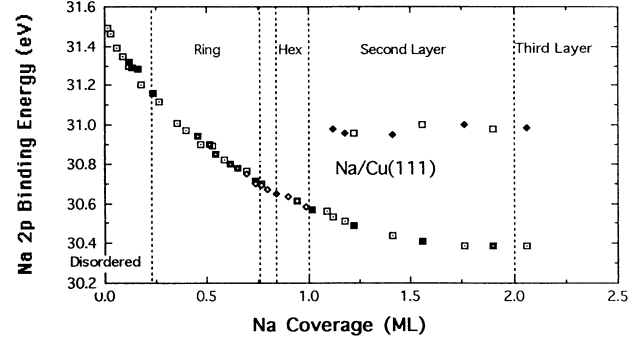


FIG. 4. Coverage dependence of the Na  $2p$  core-level binding energy for Na/Cu(111). Taken from Ref. 14.

per alkali-metal-adsorbent in units of Debye (Db),  $\theta$  is the coverage in monolayers,  $\sigma_{\text{ML}}$  is the alkali-metal adsorbent monolayer density [which is  $8.4 \times 10^{14}$  atoms/cm<sup>2</sup> for 1 ML of Na on the Cu(110) (Ref. 24) and Cu(111) (Ref. 25) surfaces], and  $\epsilon_0$  is the permittivity constant ( $= 2.655 \times 10^{14}$  Db V<sup>-1</sup> cm<sup>2</sup>).

We have found that with this relation we can fit our experimental data if we assume the following forms for the dipole moments  $D(\theta)$  of Na/Cu(110) and Na/Cu(111):

$$D(\theta)_{\text{Na/Cu(110)}} = 1.37\theta^2 - 3.42\theta + 2.66 \quad (\theta \leq 1), \quad (2)$$

$$D(\theta)_{\text{Na/Cu(111)}} = 2.73\theta^2 - 6.28\theta + 4.26 \quad (\theta \leq 1). \quad (3)$$

These coverage-dependent dipole moments are illustrated in Fig. 5(a). The polynomial nature of the functions was motivated by the theoretical calculations of Ishida of the dipole moment versus coverage for Na/jellium.<sup>26</sup> The work-function changes of Na/Cu(111) and Na/Cu(110) calculated using Eqs. (1)–(3) are presented in Figs. 5(b) and 5(c) along with our experimental data.

As tabulated in Table I, Na/Cu(110) and Na/Ni(110) exhibit more shallow minima in their work-function curves, i.e., smaller decreases from the clean value to the minimum and from the completion of the first layer to the minimum than Na/Cu(111) and Na/Ni(111). This behavior is reflected in our calculations of the dipole moments. We find that the initial dipole moment and dipole-dipole interactions are reduced in Na/Cu(110) as compared to Na/Cu(111). Our results are also in agreement with the difference exhibited by the initial dipole moments for Na/Ni(110) versus Na/Ni(111) as reported by Gerlach and Rhodin<sup>15</sup> (see Table I).

In the missing-row reconstruction model, the Na atoms are somewhat embedded in the troughs of the Cu(110) surface.<sup>27</sup> This should result in smaller dipole moments for Na/Cu(110) during the MR reconstructed phase due to a reduction in the degree of polarization of the valence charge, as compared to Na/Cu(111). With increasing coverage, we expect the mutual depolarization due to dipole-dipole interaction also to be smaller for Na in the troughs of Cu(110) versus Na atoms adsorbed on top of the Cu(111) surface. In other words, the dipole moment associated with the Na and the dipole moment decrease due to dipole-dipole interaction will both be reduced for Na/Cu(110) as compared to Na/Cu(111) due to

the different geometric configuration of the overlayer. This accounts for the different work-function behavior [i.e., the shallower minimum for Na/Cu(110)]. This discussion also applies to the differences in Na/Ni(110) versus Na/Ni(111).<sup>15</sup> Similar explanations were used to account for the work-function behavior in studies of

Cs/Ag(110) at 100 K versus 300 K by Döhl-Oelze, Stuve, and Sass<sup>1</sup> and Na/Ni(110) at 100 K versus 360 K by Memmel *et al.*<sup>4</sup> At 100 K in both cases, the reconstruction was suppressed, and the work-function minimum was deeper than at 300 K (360 K), at which temperature the (1×2) reconstruction was observed.

### B. Core-level photoemission

Turning now to our core-level results, we have also detected a qualitative difference in the Na 2*p* binding-energy versus coverage curve for Na/Cu(110) and Na/Cu(111). At higher Na coverages on Cu(110) (greater than 0.2 ML), the Na 2*p* core level decreases monotonically in binding energy and is similar in behavior to our previous results of Na/Cu(111). There is, however, a small difference in net binding-energy change over this high coverage range: ~0.45 eV for Na/Cu(110) versus ~0.65 eV for Na/Cu(111) from 0.2–1.0 ML.

Based on a comparison of core-level measurements of Na/Cu(111) and Na/Ni(111) with a theoretical calculation of Na on Jellium,<sup>14</sup> we have shown that the overall binding-energy decrease detected for these systems (though not the detailed shape) is due to an initial-state shift in the electrostatic potential energy around the alkali-metal core which is associated with a coverage dependence of the valence charge distribution and, in particular, with the degree of polarization of the alkali-metal valence charge towards the surface. For high alkali-metal coverages, the valence charge is rather symmetrically distributed about the Na nucleus. As the coverage decreases, the charge becomes more and more polarized towards the surface. This changes the potential energy at the core, resulting in an initial-state shift in the core-level binding energy (a shift to higher binding energy with decreasing coverage) and also leads to a coverage dependence in the alkali-metal dipole moment, as discussed above.

For the case of Na/Cu(110), at saturation coverage the surface is covered by a dense Na overlayer very similar to the case of Na/Cu(111). Not surprisingly, the core-level binding energy is identical at this point for the two cases. For lower coverages, the valence charge will become more polarized towards the surface, resulting in an increase in the core-level binding energy as for Na/Cu(111) and Na/Jellium. However, along with the decrease in the Na density for Na/Cu(110), there occurs a “reappearance” of Cu surface atoms which, due to the nature of the reconstruction of the Cu(110) surface, will be intermixed in the Na overlayer. Their presence within the Na layer will tend to moderate the extent of the alkali-metal valence charge polarization. This could account for the slightly smaller overall binding-energy change observed for Na/Cu(110) versus Na/Cu(111) over the high coverage range, as pointed out above.

As the Na coverage is further reduced, at some point the long-range ordering of the reconstruction will be lost [the (1×2) LEED pattern disappears at ~0.2 ML] and the Na atoms embedded in the copper surface will be fairly well isolated, as confirmed by the STM results of

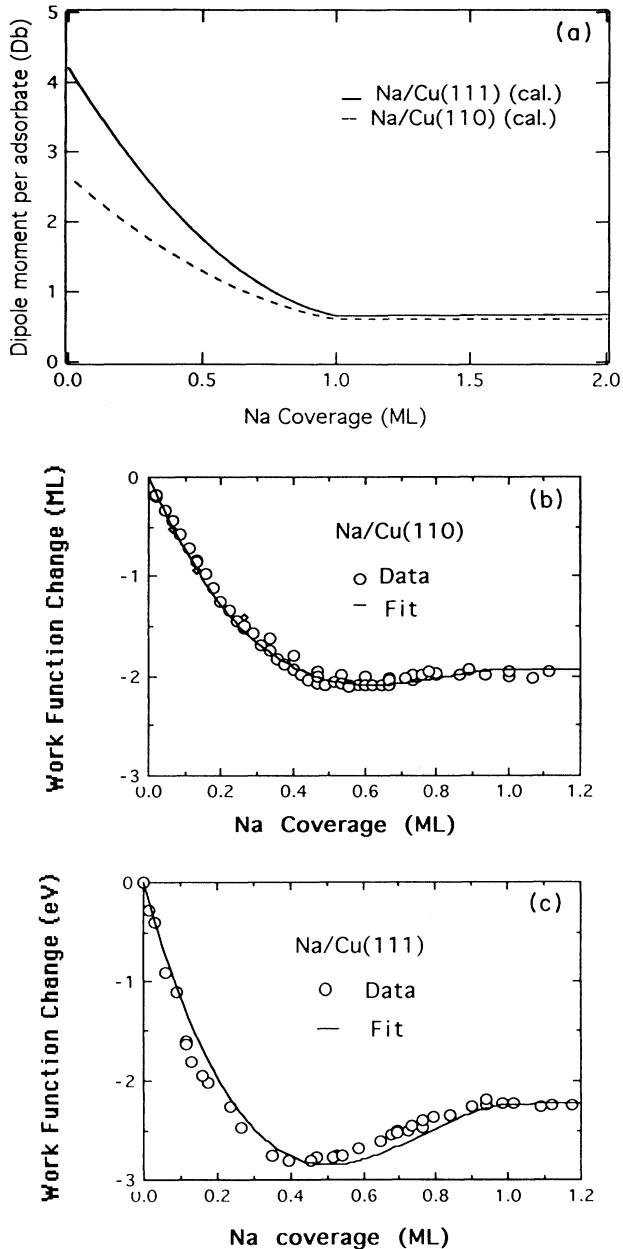


FIG. 5. (a) The dipole moment per adsorbate  $D(\theta)$  as a function of Na coverage derived from a fit to work-function data. See Eqs. (1)–(3) in the text. The dashed line is  $D(\theta)$  for Na/Cu(110), and the solid line is  $D(\theta)$  for Na/Cu(111). (b) Experimental data (open circles) and a model calculation (solid line) for the work-function change of Na/Cu(110) as a function of Na coverage. (c) Experimental data (open circles) and a model calculation (solid line) for the work-function change of Na/Cu(111) as a function of Na coverage. Data from Ref. 14.

K/Cu(110).<sup>12</sup> At this point, we believe the degree of valence charge polarization is saturated and will not be affected by a further decrease in Na coverage due to the isolated nature of the Na atoms. This then accounts for the leveling off of the core-level binding energy at about the same point at which we no longer observe the reconstructed LEED pattern, and also accounts for the smaller overall net change in binding energy for Na/Cu(110) versus Na/Cu(111). We should also like to point out that this plateauing could be caused by islanding of the Na, at this point, but we have no additional evidence to support this from LEED.

Our measurements further show that the Na 2*p* binding energy of Na/Cu(110) has a transition from a linear decrease to a plateau at very low coverage (less than 0.05 ML). One possibility is a change in the Na position at this point from top sites to vacancy sites, if the local reconstruction does not occur before the appearance of the plateau in the binding-energy curve. This could be due to diffusion of the Na atoms on the surface at very low coverage and/or it may be necessary to obtain a minimum charge-density difference at the surface before the reconstruction is energetically favorable.<sup>28</sup> These suggestions do not contradict the STM results of K/Cu(110) (Ref. 12) because the smallest coverage reported in the STM investigation corresponds to the plateau range in our binding-energy curve and not to the lowest coverage regime.

## V. CONCLUSION

In summary, we have studied the system of Na/Cu(110) using core-level photoemission, LEED, and work-function measurements as a function of Na coverage. We find that LEED pattern sequences and work-function behavior as a function of coverage are similar to that of other alkali-metal/fcc(110)-metal systems. Using a simple calculation, we can explain the different work-function behavior of Na/Cu(110) versus Na/Cu(111) as the result of a reduced dipole moment for Na/Cu(110) which we attribute to the embedded nature of the adatoms on this surface. We observe a low coverage plateau in the binding energy of the Na 2*p* core level as a function of coverage which we attribute to the effects of the Na-induced Cu(110) reconstruction. We interpret our results within a local reconstruction model of the surface.

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