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# Work functions for models of scandate surfaces

Wolfgang Müller *

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## Abstract

The electronic structure, surface dipole properties, and work functions of scandate surfaces have been investigated using the fully relativistic scattered-wave cluster approach. Three different types of model surfaces are considered: (i) a monolayer of Ba–Sc–O on W(100), (ii) Ba or BaO adsorbed on $Sc_2O_3$ + W, and (iii) BaO on $Sc_2O_3 + WO_3$. Changes in the work function due to Ba or BaO adsorption on the different surfaces are calculated by employing the depolarization model of interacting surface dipoles. The largest work function change and the lowest work function of 1.54 eV are obtained for Ba adsorbed on the Sc–O monolayer on W(100). The adsorption of Ba on $Sc_2O_3$ + W does not lead to a low work function, but the adsorption of BaO results in a work function of about 1.6–1.9 eV. BaO adsorbed on $Sc_2O_3 + WO_3$, or scandium tungstates, may also lead to low work functions.

## 1. Introduction

Several different models have been suggested for the surface structure and improved emission of scandate cathodes. Many researchers believe that a Ba–Sc–O surface layer forms when tungsten is mixed with either scandium oxide or scandium tungstate and the cathode is activated with the impregnant. In the 'top layer' cathode of Hasker et al. [1] the emission is believed to come from a Ba–Sc–O complex that forms at the $Sc_2O_3$ + W surface. Deckers et al. [2] found that W and Sc are strongly bound together and that Ba and O adsorb preferentially at these sites. Yamamoto [3] observed that a partial oxidation of the $Sc_2O_3$ + W substrate is beneficial, and he replaced the scandia by a scandium tungstate, $Sc_2W_3O_{12}$. With this configuration, less activation time was required and more consistent emission properties were obtained. During activation, he proposed that the scandium tungstate reacts with free Ba to produce free Sc, which is then available to form a Ba–Sc–O monolayer at the surface. Forman and Lesny [4] suggested that scandate cathodes operate more like oxide cathodes, rather than like activated metal surfaces in dispenser cathodes. They proposed that BaO on $Sc_2O_3$ is responsible for the improved emission and that W merely provides a conducting path for the electrons.

In this paper, models for some of the above surface structures are investigated with methods of computational quantum chemistry. Surface dipole properties are calculated and work function curves are then derived for different systems. This approach has provided excellent results for dispenser cathodes, and a mechanism has been suggested for the improved emission from M-type (tungsten-alloy) versus B-type (tungsten) cathodes [5]. The improved emission was attributed to the smaller depolarization of

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the surface dipole and the resulting larger work function change and lower final work function for the hexagonal alloy substrates relative to the cubic tungsten substrates.

## 2. Computational method

The electronic structure and charge distribution at the surface of different models for scandate cathodes has been investigated using the fully relativistic scattered-wave cluster approach, which has been described previously [6]. By use of cluster calculations, surface dipole moments can be obtained for low and high coverage conditions. The depolarization of the dipoles at higher coverage is used to calculate the polarizability of the dipole network. The initial surface dipole and the polarizability are then utilized to determine entire work-function/coverage curves.

The calculated change in the surface dipole $\mu$ due to adsorbate interactions,

$$\Delta\mu = \mu_0(\text{initial}) - \mu_f(\text{final, per adsorbate}),$$

is related to the polarizability $\alpha$ by

$$\Delta\mu = \frac{\alpha\mu_0}{j} \cdot \sum_i R_i^{-3}.$$

Here $j$ is the number of interacting dipoles, and the sum runs over distances $R_i$ from all dipoles to all other dipoles. $R_i$ is defined as $(r_i^2 + r_z^2)^{1/2}$, where $r_i$ is the separation between the dipoles and $r_z$ is one half of the dipole length of the initial surface dipole, or $\mu_0/2q_0^{\text{Ba}}$, with $q_0^{\text{Ba}}$ being the initial net charge on Ba.

For a centered square network of 5 interacting dipoles, which is used for the present investigation, $\alpha$ is obtained as

$$\alpha = \frac{5\Delta\mu}{8\mu_0} \frac{R_s^3 \cdot R_l^3}{R_s^3 + R_l^3}.$$

$R_l$ is related to the longer separation $r_l$ between the dipoles on the square network and $R_s$ is related to the shorter separation $r_s$ with respect to the dipole at the center.

Theoretical work-function curves $\phi(n)$ are derived from the depolarization model according to [7]

$$\phi(n) = \phi_0 - \Delta\phi = \phi_0 - 1.88e\mu_0 n/(1 + c\alpha n^{3/2}),$$

where $n$ is the adsorbate coverage (in $10^{15}$ atoms/cm$^2$), $\phi_0$ the initial work function, $e$ the electronic charge, $\mu_0$ the initial surface dipole, and $\alpha$ the polarizability. The constant $c$ is dependant on the geometry of the dipole network and the cluster type, and is adjusted for one reference system. A value of $c = 45$ is used for the present calculations.

## 3. Scandate surface models

The scandate surface has been modeled with Ba or BaO adsorbed on Sc, O, and W. Three different types of surfaces have been investigated: (i) a monolayer of Ba–Sc–O on W(100), (ii) Ba or BaO adsorbed on $Sc_2O_3$ + W, and (iii) BaO adsorbed on $Sc_2O_3 + WO_3$.

For the monolayer of Ba, Sc, and O on W(100), the tungsten substrate is represented by a $W_{25}$ cluster with 16 (4 × 4) surface W atoms and 9 (3 × 3) subsurface W atoms. Four Sc atoms and five O atoms are adsorbed at alternate fourfold-hollow sites on $W_{25}$, and Ba is adsorbed on top of O. One or five Ba atoms are used for modeling low and high coverage conditions, respectively. The surface structure of the resulting $Ba_{1,5}/Sc_4O_5/W_{25}$ cluster is shown in Fig. 1. The heights of the atoms above the W(100) surface are: 1.5 $a_0$ for O, 3.25 $a_0$ for Sc, and 6.5 $a_0$ for Ba (1 $a_0$ = 0.5292 Å).

Ba or BaO adsorbed on a mixed scandia–tungsten surface is modeled by using a $Sc_8W_4O_{13} = (Sc_2O_3)_4W_4O$ substrate cluster. For this surface the general structure of the $W_{25}$ cluster is maintained.

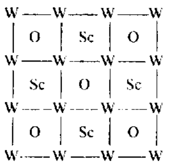

Fig. 1. Cluster model for Sc–O monolayer on W(100). Ba is adsorbed on top of O.

The 16-atom surface layer is composed of 4 Sc atoms at the center, 4 W atoms at its corners, and 8 O atoms at the remaining sites. The 9-atom subsurface layer consists of 1 O atom at its center, 4 Sc atoms as its nearest neighbors, and 4 O atoms as next nearest neighbors. For adsorbed Ba and O, heights of 6.0 $a_0$ and 1.5 $a_0$ are used, respectively, and for Ba alone a height of 4.0 $a_0$ is used.

In order to study the effect of additional oxidation, a $Sc_8W_4O_{21} = (Sc_2O_3)_4(WO_3)_3W$ scandium tungstate cluster is investigated. Here 8 Sc atoms occupy the center and the corners of the surface layer, and 8 O atoms the remaining sites. The subsurface layer contains 1 O atom at its center, 4 W atoms as nearest neighbors, and 2 O atoms in each diagonal direction. Four additional O atoms are adsorbed above the subsurface W atoms, and BaO is adsorbed at the remaining sites. Ba and O heights above the surface of 6.0 $a_0$ and 1.5 $a_0$ are used, respectively.

The above clusters have been chosen because their composition is reasonably close to actual materials that are possible candidates for scandate cathode surfaces. The Ba–Sc–O monolayer has a Ba:Sc composition similar to that in the $Ba_3Sc_4O_9$ barium scandate, which was used as an impregnant in early scandate cathodes [8]. The Sc:W ratio of 2:1 in the $Sc_8W_4O_{13}$ substrate cluster is the same as in the $Ba_3Sc_2W_1O_9$ barium scandium tungstate, which may form by a solid state reaction during activation. And lastly, the composition in the $Sc_8W_4O_{21}$ cluster of Sc:W:O = 2:1:10.5 is comparable to that in the $Sc_2W_3O_{12}$ scandium tungstate employed by Yamamoto [3].

## 4. Results and discussion

The calculated surface dipole properties and work-function data are presented in Table 1. Results are given with and without consideration of the reference dipoles for the clusters before Ba or BaO adsorption. The calculations with consideration of the reference dipole are considered more accurate, unless the correction is so large that it causes the calculated depolarization to become numerically unreliable.

The value for the initial work function of Sc–O/W(100), $\phi_0 = 2.9$ eV, is taken from Kultashev et al. [9]. For the calculations representing the $Sc_2O_3$ + W surface the same value is used, and for the scandium tungstate a slightly smaller value of 2.8 eV is employed. The latter is taken from a report by Hill and Magnus [10] who measured this effective work function for the $Sc_2W_3O_{12}$ scandium tungstate, while those for $Sc_6WO_{12}$ and mixtures of scandium tungstates with 10% W were found to be about 0.3 eV lower.

For the Ba–Sc–O monolayer on W(100), a work-function change of −1.2 to −1.4 eV is obtained which leads to a minimum work function of 1.54 eV (with consideration of $\mu_{ref}$). This is in excellent agreement with experimental values for scandate cathodes, which range from 1.5 to 1.6 eV. The calculated Ba coverage at the work function minimum ($n_{min}$), however, is quite small. If such a low work function is actually obtained at these coverages is not known, but this result is consistent with theoretical data for other systems where Ba is adsorbed on top of O, and it would be beneficial for

Table 1
Calculated initial and final surface dipoles $\mu_{0,f}$ (D), net initial charges $q_0^{Ba}$ ($e$) on Ba, polarizabilities $\alpha$ (Å$^3$), work function $\phi$ data (eV), and optimum coverages $n_{min}$ ($10^{15}$ atoms/cm$^2$)

| System | $\mu_0$ | $\mu_f$ | $q_0^{Ba}$ | $\alpha$ | $\phi_0$ | $\Delta\phi$ | $\phi_{min}$ | $n_{min}$ |
|---|---|---|---|---|---|---|---|---|
| $Ba_{1.5}/Sc_4O_5/W_{25}$ | 20.5 | 16.5 | +1.65 | 1.5 | 2.90 | −1.23 | 1.67 | 0.10 |
| with $\mu_{ref}$ | 19.6 | 16.3 | +1.65 | 1.2 | 2.90 | −1.36 | 1.54 | 0.11 |
| $Ba_{1.5}/Sc_8W_4O_{13}$ | 11.4 | 5.9 | +0.80 | 4.0 | ~ 2.9 | −0.36 | 2.5 | 0.05 |
| with $\mu_{ref}$ | 6.8 | 5.0 | +0.80 | 1.7 | ~ 2.9 | −0.38 | 2.5 | 0.09 |
| $(BaO)_{1.5}/Sc_8W_4O_{13}$ | 24.4 | 17.7 | +1.39 | 2.8 | ~ 2.9 | −0.97 | 1.9 | 0.06 |
| with $\mu_{ref}$ | 19.9 | 16.8 | +1.39 | 1.3 | ~ 2.9 | −1.31 | 1.6 | 0.11 |
| $(BaO)_{1.5}/Sc_8W_4O_{21}$ | 24.2 | 16.8 | +1.35 | 3.2 | ~ 2.8 | −0.88 | 1.9 | 0.06 |

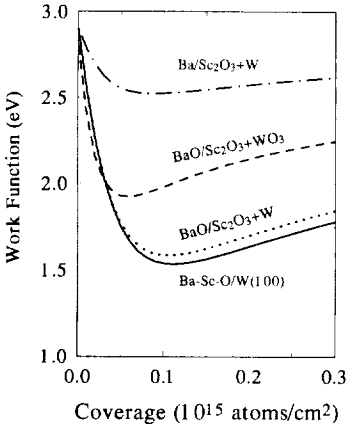

Fig. 2. Work function curves for scandate model surfaces.

cathode operation and life because a low work function could be maintained with small Ba coverages.

When yttrium is substituted for scandium in the Ba–Sc–O overlayer, very similar results are obtained for the surface dipole properties and resulting work functions. No definitive conclusion could be drawn, however, on a possible improvement due to this substitution, because $\phi_{min}$ for the Ba–Y–O overlayer as compared to the Sc system is somewhat higher (1.78 eV) without $\mu_{ref}$, but lower (1.43 eV) with consideration of $\mu_{ref}$.

For the substrate cluster representing an approximate 1:1 mixture of $Sc_2O_3$ and W, $Sc_8W_4O_{13}$, Ba adsorption gives rise to only a modest work-function change (−0.4 eV), while BaO adsorption gives rise to a much larger change (−1.0 to −1.3 eV). Based on a $\phi_0$ of 2.9 eV, the adsorption of BaO leads therefore to minimum work functions of 1.9 and 1.6 eV, without and with consideration of $\mu_{ref}$, respectively. The latter value is in agreement with the good results that are obtained with mixed scandia–tungsten matrices.

When the substrate is further oxidized, $Sc_2O_3$ + W is transformed into $Sc_2O_3 + WO_3$, or scandium tungstate. Such a surface is represented by a $Sc_8W_4O_{21} = (Sc_2O_3)_4(WO_3)_3W$ cluster. (The data given in Table 1 are those without consideration of the reference dipole because the inclusion of the large $\mu_{ref} = 8.4$ D for this cluster makes the depolarization unreliable.) Based on the quoted value of $\phi_0 = 2.8$ eV, BaO adsorption leads to a calculated work function change of −0.9 eV and a minimum work function of 1.9 eV. It is interesting to note that almost identical results have been obtained experimentally by Hill and Magnus [10] for the comparable $Sc_2W_3O_{12}$ scandium tungstate. The calculated work function curve is shown in Fig. 2 together with the results for the other scandate model surfaces. Because the $Sc_8W_4O_{21}$ cluster contains some additional W, it would also be reasonable to use a $\phi_0$ of 2.5 eV which was measured for mixed scandium tungstate + tungsten surfaces [10]. With this $\phi_0$ a minimum work function of 1.6 eV would be obtained, which is very similar to the result for BaO on $Sc_2O_3$ + W.

## 5. Summary and conclusions

Among the three types of model surfaces that have been considered for scandate cathodes, the monolayer of Ba–Sc–O on W(100) produces the lowest work function. The calculated values of 1.5–1.7 eV are in excellent agreement with experimental results for scandate cathodes. The adsorption of Ba on $Sc_2O_3$ + W does not produce a significant work-function change, but the adsorption of BaO on $Sc_2O_3$ + W leads to a work function of about 1.6–1.9 eV. It is therefore mandatory that Ba is directly bound to O, and Ba is possibly positioned directly above O. The result for BaO on $Sc_2O_3$ + W is in agreement with the high emission observed from cathodes with scandia–tungsten matrices. The adsoption of BaO on $Sc_2O_3 + WO_3$ is likely to also produce low work functions.

The investigation of scandate cathode models points at this stage to the Ba–Sc–O monolayer on W as a possible surface with the proper work function. It should be emphasized, however, that the more highly oxidized substrate clusters represent simplified models which are representative only for the appropriate compositions rather than the actual surface structures of these materials. Much more work is needed to better define the actual atomic arrangements at the surface, experimentally and/or theoretically, before a final conclusion can be reached about the true surface structure of operating scandate cathodes.

## Acknowledgements

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