

Esaki Diodes Based on 2-D/3-D Heterojunctions

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Abstract—Esaki diodes based on interband tunneling have the characteristics of negative differential resistance (NDR) and ultrafast transient time, which lead to broad applications including oscillators, multivalue memories, and terahertz detectors. In this paper, we present the first demonstration of Esaki diodes based on 2-D/3-D heterojunctions—more specifically, chemical vapor deposition MoS₂ on degenerately doped silicon. As compared to traditional 3-D heterostructures, these 2-D/3-D heterostructures have the following advantages: dislocation-free 2-D crystals even when the lattices are mismatched, dangling bond-free surface, and capability for large-scale synthesis at low cost. In this paper, monolayer, bilayer, and trilayer MoS₂ are synthesized directly on degenerated Si substrate, forming the ultraclean heterostructures without surface contamination from tape and resist. Based on these pristine heterostructures, we are able to observe prominent NDR effect at room temperature. This NDR effect is attributed to the degenerately p-type doping in silicon and the natural n-type doping in MoS₂. We also found that the peak voltage corresponding to the local maximum tunneling current depends on the MoS₂ thickness. While MoS₂ is changing from the monolayer (0.7 nm) to bulk (9.5 nm), the peak voltage increases from 0.8 to 1.6 V. This phenomenon can be explained by the energy-level differences between the monolayer and bulk MoS₂. This paper provides the experimental groundwork for the synthesis of transition metal dichalcogenides on degenerately doped Si substrates and opens up new and exciting opportunities for electronic applications of 2-D/3-D heterostructures.

Index Terms—2-D/3-D heterojunctions, chemical vapor deposition (CVD), degenerately doped silicon, Esaki diodes, MoS₂.

I. INTRODUCTION

ESAKI diodes have unique negative differential resistance (NDR) characteristics, which enable their applications in oscillators, terahertz detectors, multivalue memories, and analog-to-digital converters [1]–[3]. In addition, Esaki diodes serve as the critical components in tunneling field-effect

transistors, which have potential applications in low-power electronics [4]–[6]. Various material stacks have been used in Esaki diodes, including Si, Ge, SiGe, group III-V, and their heterostructures [7]–[13]. Among these materials, III-V heterostructures grown by molecular-beam epitaxy produced the highest performing Esaki diodes [14]. The key factors that deteriorate the performance of III-V-based Esaki diodes are lattice mismatch (and associated dislocations), impurity scattering, polarization charges, and instability of space charge [14].

Recently, 2-D crystals have emerged as promising candidates for Esaki diodes [15], [16]. 2-D materials are free of surface dangling bonds, and 2-D heterostructures bonded by van der Waals forces are free of dislocations even when there is a large mismatch in lattice constants [17]. Esaki diodes based on SnSE₂/black phosphorus have been demonstrated, where the broken-gap band alignment between these two materials enables the NDR effect [15]. However, black phosphorus is not thermodynamically stable and is difficult to be synthesized in a large scale. In contrast, transition metal dichalcogenides (TMDs) can be synthesized in large scale via chemical vapor deposition (CVD) or molecular organic CVD [18]–[25]. However, most of the 2-D TMD heterostructures, such as MoS₂/WSE₂ heterostructure, do not have broken-gap band alignment. It is reported that by applying a vertical electric field to offset the band alignment between these two materials, NDR behavior can be observed [16]. However, the NDR effect in these devices is very weak and can only be observed at low temperatures. In addition, this device requires four electrodes instead of two electrodes in traditional Esaki diodes, which adds process complexity and device footprint.

In this paper, we propose a new type of Esaki diodes based on 2-D/3-D heterojunctions. In these devices, the 3-D material, such as silicon, is degenerately doped, while the 2-D material is naturally or intentionally doped into the opposite type to form a heterogeneous p-n-junction. This 2-D/3-D heterostructure can effectively eliminate the lattice-match restriction in a 3-D/3-D structure. As compared to the 2-D/2-D structure, the 2-D/3-D structure takes the advantages of the mature silicon technology and the commercially available degenerately doped silicon substrate. The 2-D TMDs can be grown directly on the silicon substrates, which will enable large-scale low-cost fabrication of the heterostructures and avoid surface/interface contamination from the transfer process. These pristine heterostructures enable us to observe prominent NDR effect at room temperature. This NDR effect is attributed to the energy-band alignments between the n-type

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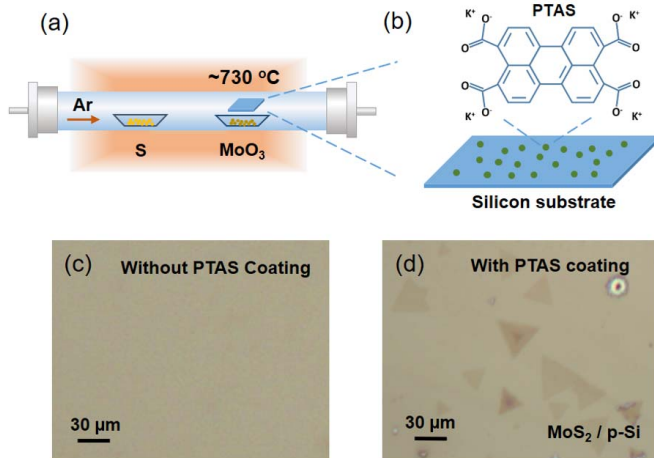


Fig. 1. Synthesis of MoS₂ on silicon substrates. (a) Schematic of the MoS₂ CVD system. (b) Chemical structure of PTAS. (c) and (d) Optical images of MoS₂ grown on degenerately doped silicon wafers without and with PTAS seeding promoter, respectively.

MoS₂ and degenerately doped p-type Si. Moreover, the tunneling transports of MoS₂/p-Si heterostructures with different MoS₂ thicknesses are also investigated. We found that the peak voltage, V_P , is dependent on the MoS₂ thickness. As MoS₂ thickness changes from 0.7 nm (monolayer) to 9.5 nm, the peak voltage increases from 0.8 to 1.6 V, which can be explained by the thickness dependence of the energy bands in MoS₂.

II. RESULT AND DISCUSSION

Recently, considerable efforts have been made to grow monolayer MoS₂ nanosheets by using CVD [22], [26]. However, most of the work focuses on the synthesis of MoS₂ on SiO₂ or sapphire substrates. Here, we systematically investigated MoS₂ synthesis on degenerately doped silicon substrates with a resistivity of 0.001 Ωcm. Fig. 1(a) illustrates the setup of the CVD system. Molybdenum oxide (MoO₃) and sulfur (S) powders were used as precursors and were placed in two separated crucibles. The two zones with MoO₃ crucible and S crucibles were heated to 730 °C and 200 °C, respectively. The degenerately doped p-type Si substrates were facing down on the MoO₃ crucible. The MoS₂ layers were synthesized for 5 min with argon flow at ambient pressure. Without seeding promoter in the silicon substrate, the yield of MoS₂ was very low and only MoS₂ particles were obtained, as shown in Fig. 1(c). To address this nucleation issue, perylene-3,4,9,10-tetracarboxylic acid tetrapotassium salt (PTAS) was used as the seeding promoter in the MoS₂ synthesis. Fig. 1(b) shows the molecular structure of PTAS. As compared to other aromatic molecules, PTAS has better thermal stability [18]. Moreover, the high solubility of PTAS in water can facilitate the uniform distribution of the seeds on the Si substrate. With PTAS as the seeding promoter, monolayer and multilayer MoS₂ nanosheets were successfully synthesized on a Si substrate, as shown in Fig. 1(d). Note that monolayer and multilayer MoS₂ coexist on the same wafer. Typically, multilayer MoS₂ is located at the center of the triangle, while monolayer MoS₂ is located at the outer zone of the triangle.

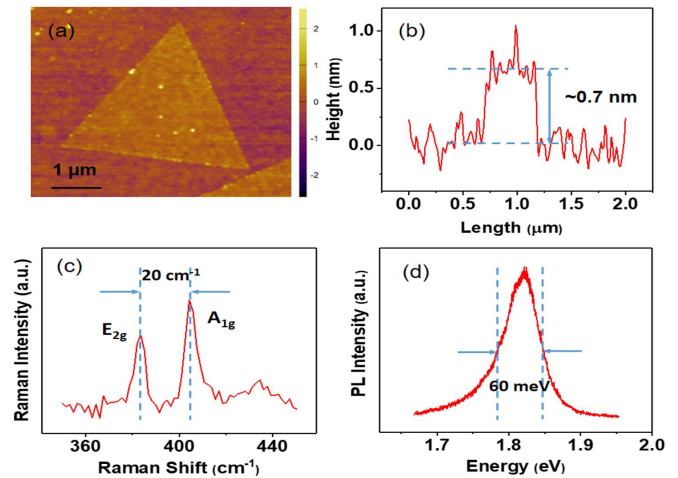


Fig. 2. Characterization of MoS₂ synthesized on silicon substrates. (a) and (b) AFM image and line profile of the MoS₂ film. (c) and (d) Raman and PL spectra of the MoS₂ synthesized by CVD, respectively.

The grain size of synthesized MoS₂ is about 30 μm. The thickness of the MoS₂ is ~0.7 nm measured by atomic force microscopy (AFM) [Fig. 2(a) and (b)], indicating that it is a monolayer. Furthermore, the synthesized MoS₂ are characterized using Raman spectroscopy and photoluminescence (PL). In the Raman spectrum shown in Fig. 2(c), there are two peaks located at 384 and 404 cm⁻¹, corresponding to E_{2g} and A_{1g} modes in the monolayer MoS₂, respectively. It has been reported that the wavenumber difference between these two peaks (E_{2g} and A_{1g}) is about 20 cm⁻¹ for monolayer MoS₂, whereas, it is 25 cm⁻¹ for bulk MoS₂ [27]. The difference between these two peaks in our synthesized MoS₂ sample is ~20 cm⁻¹, further confirming that the synthesized MoS₂ film is a monolayer. The PL peak of the MoS₂ sample is at around 1.83 eV with a full-width at half-maximum intensity of 60 meV [Fig. 2(d)], corresponding to the feature of monolayer MoS₂ with high crystal quality [22]. In the future research, growth condition is to be further optimized to realize the wafer-scale growth of MoS₂ on the Si substrate.

Conductive AFM (C-AFM) measurements were performed to investigate the tunneling transport of our CVD MoS₂/Si heterostructures. As compared to the traditional IV measurement on vertical devices with electrodes, the C-AFM measurement has several advantages, including contamination free (avoiding the lithography-induced resist residuals), nanoscale probing on the local tunneling current (minimizing the noise induced by nonuniformity of the film), fast turn-around time, and direct correlation between the topography (layer thickness) and tunneling characteristics [28]. Fig. 3(a) shows the schematic of the C-AFM setup. Here, conductive Cr/Pt coated tips were used as the local electrical probes to obtain reliable measurements. The tip radius of our C-AFM is less than 25 nm. Note that for our C-AFM measurement, the current limit is set at 20 nA.

CVD monolayer, bilayer, and trilayer MoS₂ on silicon were identified by AFM in the topography mode, as shown in Fig. 3(b). The tunneling currents were measured *in situ* in the C-AFM mode as shown in Fig. 4(a). The zoomed-in

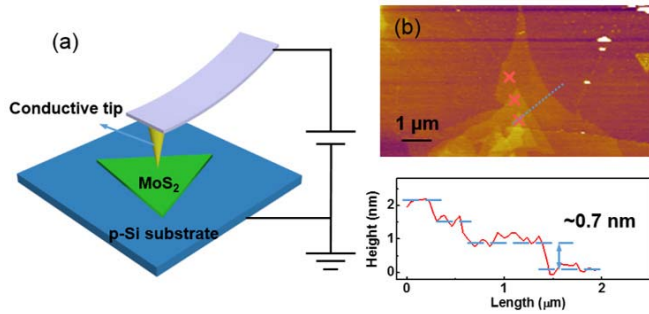


Fig. 3. (a) Schematic of the setup of C-AFM measurement. (b) AFM image and line profile of the CVD MoS₂ on a silicon substrate.

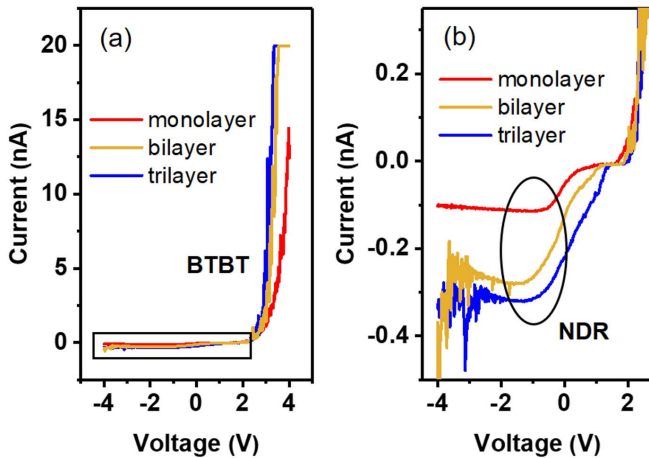


Fig. 4. C-AFM measurements of CVD MoS₂ on degenerately doped silicon. (a) Tunneling current as a function of voltage for monolayer, bilayer, and trilayer MoS₂ grown on silicon substrate. (b) Zoomed-in view image of the IV curves in the negative bias region marked in the box on (a).

view of the negative bias regime is exhibited in Fig. 4(b). The IV curves show a nonlinear behavior. Since the voltage is applied on n-type MoS₂, the negatively applied voltage means a forward bias on the p-n-junction. We can see that NDR behavior appears in the forward bias region. For the monolayer MoS₂/Si heterostructure, the peak voltage (i.e., the voltage when the tunneling current reaches the local maximum), V_p , is ~ 0.8 V. The peak-to-valley current ratio of the bilayer MoS₂/Si heterostructure is ~ 1.16 . This peak-to-valley current ratio is higher than that of MoS₂/WSe₂ heterostructure at room temperature but lower than that of SnSe₂/black phosphorus heterostructure [15], [16].

This NDR effect can be explained by the energy-band alignment between the degenerately doped silicon and naturally doped n-type MoS₂. Fig. 5(b)–(e) show the energy diagrams of MoS₂/p-Si heterostructure at various bias conditions and Fig. 5(a) illustrates the corresponding tunneling current at these bias conditions. When a reverse bias is applied on the heterostructure (i.e., a positive voltage is applied on the n-type MoS₂), the electrons from the valence band of Si tunnel into the conduction band of MoS₂ and the tunneling current increases exponentially with increasing reverse bias [Fig. 5(b)]. The corresponding current is designated by a

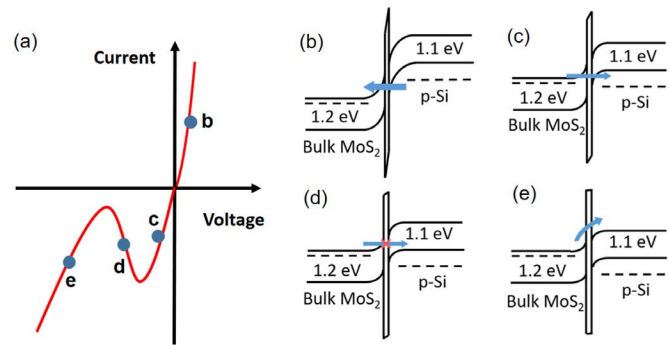


Fig. 5. Energy diagrams of MoS₂/Si heterojunctions at various bias conditions. (a) Illustration of the IV curves with NDR effect. Four different regions are marked as b–e. The corresponding energy diagrams are illustrated in (b) reverse bias, (c) small forward bias with band-to-band tunneling current flowing, (d) forward bias such that the tunneling current is prohibited, and (e) high forward bias with thermal current flowing.

circle “b” on the IV curve [Fig. 5(a)]. When a small forward bias is applied, a band of energies exists for which there are filled states on the n-type MoS₂ side, while unoccupied states on the p-type Si, thus the electrons can tunnel from n-MoS₂ to p-Si [Fig. 5(c)]. When the forward bias is further increased, there are fewer unoccupied available states on the p-Si side, which allows the tunneling of electrons from the n-MoS₂ side. When the decrement of the tunneling current due to reduced available states is dominant over the increment of the tunneling current due to an increased electric field across the junction, the tunneling current starts to decrease. As the forward bias is further increased such that the conduction band edge of the n-type MoS₂ matches or exceeds the valence band edge of the p-type Si, electrons in MoS₂ can no longer tunnel into a valence band state in silicon [Fig. 5(d)]. With still further increase in the bias, the electron and hole diffusion barrier will reduce and the thermal current will start to increase, therefore, the total current will increase again [Fig. 5(e)]. The decreasing portion of the tunneling current after the peak current [region d in Fig. 5(a)] gives rise to the NDR effect. Note that typically 2-D MoS₂ grown by the CVD method is naturally n-type doped [19], [20]. To create Esaki diodes, degenerately doped p-type Si substrates were used. At equilibrium, if the conduction band edge of MoS₂ in the neutral region is lower than the valence band edge of silicon in the neutral region, NDR effect can occur. Here, we assume that there is a thin layer of native silicon oxide at the interface between MoS₂ and silicon. It is reported that the thickness of the native oxide on a silicon wafer is typically ~ 1 nm at room temperature [29]. The traps in the native silicon oxide layer and the dangling bonds of the silicon surface can result in trap-assisted tunneling and degrade the peak-to-valley ratio.

To further investigate the thickness-dependent tunneling transport of MoS₂/Si heterostructures, thicker MoS₂ nanosheets are mechanically exfoliated on degenerately doped p-type Si substrate. The thickness ranges from 1.8 to 9.5 nm as confirmed by AFM, as shown in Fig. 6(a). Then, the local IV curves are measured by C-AFM with the bias from 4 to -4 V, as shown in Fig. 6(b). The zoomed-in view of the negative bias

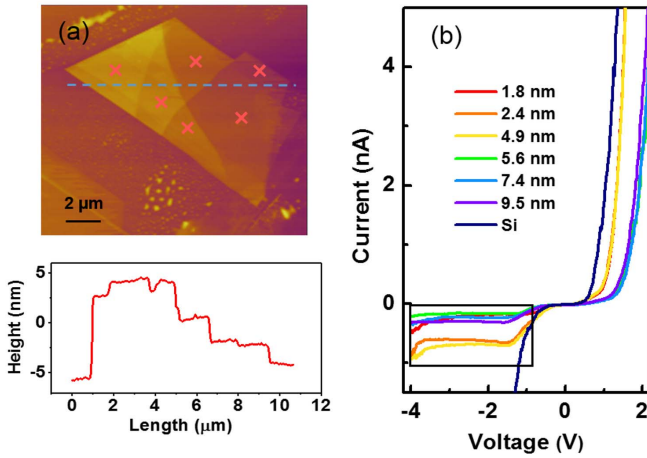


Fig. 6. C-AFM measurements of exfoliated MoS₂ on a degenerately doped p-type silicon substrate. (a) AFM image and line profile of the MoS₂ on silicon. (b) Tunneling current as a function of applied voltage for MoS₂ with various thicknesses.

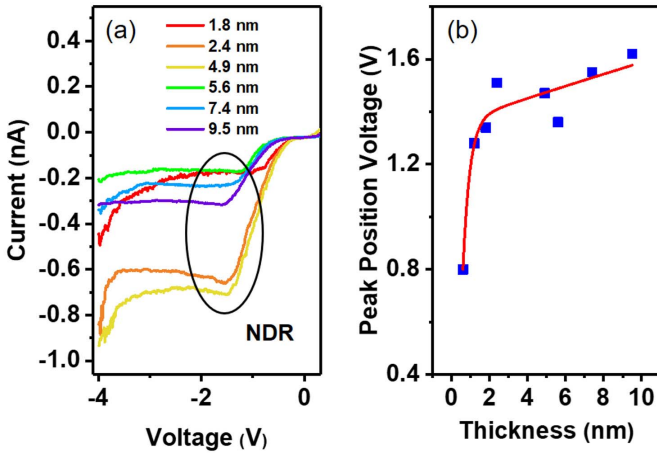


Fig. 7. Thickness dependence of the peak voltage. (a) Zoomed-in view of the tunneling current as a function of applied voltage in the negative bias regime for MoS₂ with various thickness. (b) Peak voltage as a function of MoS₂ thickness. Blue squares: measured data. Red line: guiding eyes only.

regime is shown in Fig. 7(a). Importantly, the NDR behaviors are clearly observed in MoS₂/Si heterostructures with various MoS₂ thicknesses. In contrast, there is no NDR effect observed in the bare Si sample without MoS₂. This indicates that the NDR effect observed in MoS₂/Si samples is related to the MoS₂ layer. Furthermore, we found that the peak voltages are dependent on the MoS₂ thickness. As shown in Fig. 7(b), as the MoS₂ thickness increases from 0.7 nm (monolayer) to 9.5 nm (bulk), the peak voltage increases from 0.8 to 1.6 V. This can be explained by the energy-level differences between the monolayer and bulk MoS₂. The peak-to-valley ratios for different MoS₂ thicknesses range from 1.2 to 1.05, with no strong dependence on the film thickness. The IV curves show nonzero current at zero voltage, which results from the background noise current at tens of picoampere.

Fig. 8(a) shows the energy diagram of the isolated monolayer MoS₂, bulk MoS₂, and degenerately doped p-type

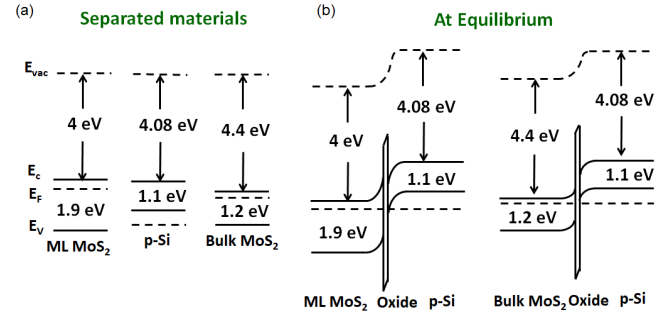


Fig. 8. Energy diagrams of MoS₂/Si heterojunctions. (a) Energy diagram of isolated monolayer MoS₂, bulk MoS₂, and degenerately doped silicon before they contact each other. (b) Energy diagrams of monolayer-MoS₂/Si and bulk-MoS₂/Si heterostructures.

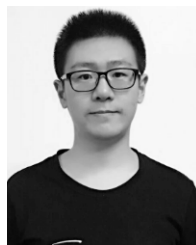
silicon. Fig. 8(b) shows the energy diagram of monolayer-MoS₂/Si and bulk-MoS₂/Si heterostructures. Based on the theoretical calculations, the electron affinity of MoS₂ is 4 eV for monolayer and 4.4 eV for bulk. Experimentally, it is reported that the work function of bilayer MoS₂ is about 0.15 eV lower than that of six-layer MoS₂ [30]. As the thickness of MoS₂ increases from a monolayer to bulk, if the work function increment is less than the electron affinity increment, then the energy difference between the conduction band edge and the Fermi level in the MoS₂ neutral region, $E_{c-\text{MoS}_2} - E_F$, will decrease. This decrease means that the energy difference between the valence band edge in Si and the conduction band edge in MoS₂ in the neutral region, $E_{v-\text{Si}} - E_{c-\text{MoS}_2}$, will increase, leading to an increase in the peak voltage as the thickness of MoS₂ increases. Note that in these measurements, the true voltage dropped on the heterojunction is less than the applied voltage, due to the voltage consumed on the tip/MoS₂ contact, MoS₂ neutral region, interfacial oxide layer, and/or the air gap between the tip and the sample.

III. CONCLUSION

In summary, we have first demonstrated Esaki diodes based on 2-D/3-D heterojunctions: CVD MoS₂ on degenerately doped silicon substrates, with prominent NDR effect at room temperature. Monolayer MoS₂ nanosheets are successfully synthesized on the degenerately doped p-Si substrate directly by CVD using PTAS as the seeding promoter. AFM, Raman, and PL characterizations confirmed that the synthesized MoS₂ is a monolayer with high crystal quality. Furthermore, C-AFM is carried out to investigate the tunneling transport of MoS₂/Si heterostructures. The NDR effect is observed in the forward bias regime, which can be explained by the band alignment between MoS₂ and degenerately doped Si. We also found that the peak voltage is dependent on the MoS₂ thickness. When the MoS₂ thickness increases from 0.7 to 9.5 nm, the peak voltage increases from 0.8 to 1.6 V. This phenomenon is attributed to the difference in the energy levels in the monolayer and bulk MoS₂. This paper will provide a new path for Esaki diodes based on synthesized 2-D/3-D heterostructures. This new type of the device has the advantages of high performance and large-scale manufacturability with low cost, giving it broad applications in electronics and optoelectronics.

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