Comment on "Quantum Contact in Gold Nanostructures by Scanning Tunneling Microscopy"

In a recent Letter by Pascual *et al.* [1], the authors applied voltage pulses to a Au scanning tunneling microscope (STM) tip to form mounds on a Au surface, in a manner similar to that of Mamin, Guethner, and Rugar [2,3]. They found that tip-sample resistances of a few hundred ohms occurred during mound formation, implying physical contact. They then inferred that this physical contact, triggered by the voltage pulse in some manner not fully understood, caused mound formation as the tip retracted. They state that "contact between tip and sample is the main requirement" for mound deposition and conclude that the field-induced atomic emission mechanism proposed by us is unlikely.

We certainly agree that such tip-sample contact often takes place. Indeed, this contact behavior has been observed by a number of groups [4,5], including ours, and has been discussed explicitly in the literature by McBride and Wetsel [6]. We would argue, though, that it can be difficult to determine whether the contact is the *cause* of the mound formation or rather a *consequence* of it. It seems equally plausible, for example, that material deposited from the tip onto the surface fills up the gap, causing a bridging contact to occur [7]. We cite below two pieces of evidence which show that material transfer can occur during STM voltage pulses without requiring contact.

First, it is well known that when refractory metal tips (e.g., tungsten) are used on a Au surface, voltage pulses can result in pits as well as mounds [4]. As shown previously by Li *et al.* [4] and McBride and Wetsel [6], in pit formation the large currents corresponding to bridging contact are *not* observed. The resistances are typically 10^6–10^7 Ω, so that there is no physical contact. Of course, it is possible that entirely different mechanisms are responsible for pit formation and mound formation. However, both processes show the same sharp threshold behavior, with similar threshold values, strongly suggesting that they are different manifestations of the same material transfer process.

In addition, we have direct evidence, using ultrashort voltage pulses, that mound formation is possible without contact. Using an impedance-matched microwave stripline sample holder and 900 ps pulses [8], we have created mounds a few Å high without the high currents associated with contact. Figure 1 shows the current as a function of time for a pulse applied at $t=0$. Immediately after the pulse, the current increased to 7 nA, corresponding to a partial filling in of the gap. The current then slowly recovered to its original value of 0.2 nA as the tip was pulled back by the servo. The minimum tip-sample resistance was 10^7 Ω, a value well in the tunneling regime. Therefore not only is contact typically absent in *pit* formation, but it is also not a necessary condition for *mound formation,* at least for sub-ns pulses.

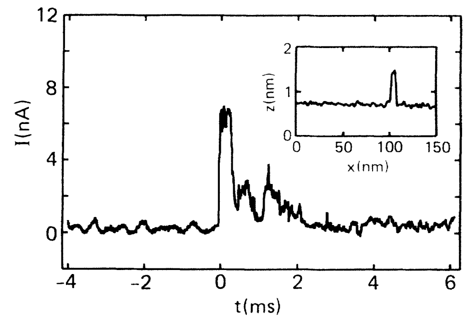

FIG. 1. Tunnel current vs time for a 5 V, 900 ps voltage pulse applied at $t=0$. The current reached a maximum of only 7 nA after the mound formation, showing that electrical contact did not occur. The inset shows a cross section of the deposited mound as measured by a subsequent STM line scan.

The authors rightfully draw attention to the issue of tip-sample contact, which may well play an important role in determining the final shape of the mound and in reforming the tip. However, such contact does not appear to be the underlying cause of the mound formation. While some open questions remain, a field-induced transfer mechanism is quite consistent with the results, and also can explain in a natural way both pit and mound formation.

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[7] Tip shape and size will change as material is transferred from it. If the reduction in tip length is less than the height of the deposited mound, contact will result. This could occur if material is transferred from a larger area into a smaller area, or if the tip sharpens in the high field.
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