Slope Water Current over the Laurentian Fan on Interannual to Millennial Time Scales

L. D. Keigwin and R. S. Pickart

The strength and position of surface and deep currents in the slope water south of Newfoundland are thought to vary as a coupled system in relation to the dipole in atmospheric sea level pressure known as the North Atlantic oscillation (NAO). Paleoceanographic data from the Laurentian Fan, used as a proxy for sea surface temperature, reveal that surface slope waters north of the Gulf Stream experienced warming during the Little Ice Age of the 16th to 19th centuries and support the notion of an NAO-driven coupled system. The NAO may be a useful model for millennial-scale ocean variability during interglacial climate states.

North of the Gulf Stream, a filament of water runs eastward over the continental margin of Newfoundland, coincident with a sharp front in temperature and salinity. This slope water front and current originates near 60°W (Fig. 1), apparently the result of a minor bifurcation of the Gulf Stream (*1*). Beneath the slope water current reside the Labrador Sea water and Denmark Strait overflow water, which flow equatorward in the deep western boundary current (DWBC). These different currents appear to act as a coupled system, and their position and intensity vary on interannual to decadal time scales (*2*). Pickart *et al*. (*2*) have suggested that this system oscillates between two modes, in a manner consistent with observed linkages between ocean convection and air-sea forcing in different regions of the North Atlantic (3). The predominant low-frequency atmospheric circulation pattern over the subpolar North Atlantic is that associated with the North Atlantic oscillation (NAO), defined as the wintertime sea-level pressure difference between Iceland and the Azores (*4*). During the minimum phase of the NAO, convection in the Greenland Sea and the Sargasso Sea is enhanced (3), which may lead to increased transport of Denmark Strait overflow water in the DWBC (*5*). The results of Pickart *et al*. (*2*) suggest that the slope water current also strengthens and moves northward during minima in the NAO, but this interpretation is more speculative. We present here paleoceanographic data from the Laurentian Fan, extending back through the Little Ice Age (LIA), that support variability in the slope water current that is consistent with the coupled slope water mode. These observations show that phasing of the sea-surface temperature (SST) on millennial time scales between the Laurentian Fan and the subtropics is the same as for NAO-related changes.

During July 1998, the R/V *Oceanus* sampled various locations on the Laurentian Fan (Fig. 1), where sedimentation rates are high because of sediment supply from the Gulf of St. Lawrence and Newfoundland margin as well as equatorward advection by the DWBC. Because the Laurentian Fan is dissected by several channels, each bearing sediment from different localities, slightly different sedimentation histories are expected for different core sites (*6, 7*).

We focused on two multicore (MC) (*8*) sites, one from the western levee of the eastern valley of the fan (MC-13) (3440 m) and one to the east of the eastern valley (MC-25) (3890 m) (Fig. 1). Each site contains bomb ^{14}C in planktonic foraminifera at the core top (Table 1), indicating that sedimentation rates are high enough that this signal, which peaked in the atmosphere in 1963, has not been mixed out by burrowing activity on the seafloor. Therefore, these sites have high enough temporal resolution for comparison to the only other site, on the northeastern Bermuda Rise (Fig. 1), where bomb ^{14}C in foraminifera is also distinct on the seafloor (*9*). We determined the calcium carbonate content as an inverse measure of input of terrigenous clays and silts, which accounts for the high rates of sedimentation (*10*), and we measured planktonic foraminiferal abundance changes as a proxy for SST.

At each location the carbonate content is low near the core top and at a second interval deeper in the cores, but the records differ in detail. The upper 2 to 5 cm of MC-13 are red, similar to glacial maximum sediments along the southeastern Canadian margin (*11*), but the top of MC-25 has a more typical brown oxidized layer. Whereas the carbonate content decreases gradually in the upper 10 cm of MC-25 (Fig. 2B), in MC-13 it drops abruptly to lower values and recovers at the core top (Fig. 2A). Because these cores have modern tops, they must be recording different local sedimentary events over the past several centuries as well as a regional signal. MC-25 has an average sedimentation rate (*12*) of about 28 cm per thousand years, roughly twice that of MC-13 (16 cm per thousand years). Thus, MC-25 should have the higher resolution record. However, the red sediment at MC-13 may have been deposited rapidly if it represents the fine-grained tail of multiple turbidites that flooded over the western levee of the eastern valley. Turbidites in the eastern valley of the fan might not affect a more easterly location such as MC-25 because the prevailing flow of the DWBC is westward. When the carbonate records of the two sites are plotted together versus age, it is evident that each site experienced the same decrease starting in the LIA about 400 years ago, and they both show the same carbonate minimum at 1200 to 1600 years before present (B.P.) (Fig. 2C). If the fine-grained red sediment near the top of MC-13 was deposited rapidly and diluted the pelagic carbonate sediment, then that core may have a better preserved record during the LIA than MC-25.

For a paleotemperature proxy, we counted the abundance of the left-coiling form of *Neogloboquadrina pachyderma* as a percentage of the total planktonic foraminiferal fauna >150 μm (Fig. 2, D and E). This species is the only polar planktonic foraminifer that appears in waters colder than about 10°C and displays a linear increase in abundance with decreasing temperature, reaching 100% of the planktonic fauna in near-freezing waters (*13*). Each core shows that *N. pachyderma* abundance decreased abruptly about 400

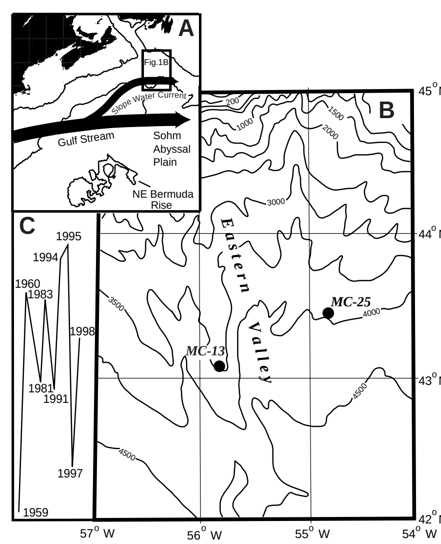

Fig. 1. (**A**) Western North Atlantic, showing the Bermuda Rise in the Sargasso Sea, the average position of the Gulf Stream, the slope water current, and inset of the Laurentian Fan (**B**). MC locations discussed in the text are shown on the Laurentian Fan and the continental slope off Newfoundland. (**C**) Historical north-south movements of the slope water current in the range 42 to 44°N, from published data (*2*), from *Oceanus* 326 data (1998), and from R/V *Knorr* 151-3 data (1997) adjusted to longitude 55°W.

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years ago (implying a warming of surface waters), coincident with the decrease in carbonate content near the core top (Fig. 2F). This pattern is unexpected because carbonate-poor intervals in North Atlantic sediments normally are associated with cooler climate (*14*). Near the top of MC-13, a warming of about 2 ± 1°C is indicated by the 20% decrease in *N. pachyderma* abundance during the LIA interval based on the abundance-SST relationship of Kohfeld *et al*. (*13*). However, unlike in MC-25, a full return to lower SST during the last century is not recorded in MC-13. This difference is probably an artifact of bioturbation and changing sedimentation rates at MC-13, as discussed above. If the red sediment was deposited rapidly and the more typical gray-brown core top sediments more slowly, then the red sediments would more accurately reflect the warming registered by *N. pachyderma* during the LIA. By contrast, the overlying 0- to 2-cm interval at MC-13 would record a more attenuated cooling during the last century (increase in percent of *N. pachyderma*) because bioturbation might have mixed some of the LIA specimens upward. It is impossible to support this assertion with ^{14}C data because calendar ages cannot be calculated for samples above 3.5 cm where the conventional ^{14}C age is <440 years (Table 1). MC-25 shows evidence of earlier warm events about 1100 and 1500 years ago, whereas the more noisy results at MC-13 may be a result of bioturbation. Although the small differences among the SST proxy data at these sites are curious, the more striking observation is that both sites indicate that slope waters must have warmed during the LIA.

We propose that the slope water current is responsible for warming this part of the North Atlantic by moving northward during the LIA. When the data from Pickart *et al*. (*2*) and from two more recent cruises are plotted in a geographic coordinate system and adjusted to the common meridian of 55°W, it is clear that the slope water current repeatedly traversed this region in the past few decades (Fig. 1C). A composite temperature section near 55°W shows that the core of the slope water current falls within the 8° to 12°C range (Fig. 3). Therefore, a northward shift in its position could cause a 4°C SST warming and a relatively large faunal change by excluding cold water fauna such as *N. pachyderma* (restricted to SSTs ≤10°C). If the surface North Atlantic cooled slightly during the LIA [evidence suggests that the subtropical gyre SST decreased 1° to 2°C during this period (*9*)], then movement of the slope water front past the core sites could result in local warming of SST over the Laurentian Fan by 1° to 2°C (the difference between the basin-wide cooling and local warming due to the frontal shift). This is the magnitude suggested by the *N. pachyderma*.

We have no evidence that the slope water current has ever resided north of 44°N, and only once (in 1995) has it approached that latitude. Core sites to the north should have colder fauna, and data from MC-19 and -21 (Fig. 1) support this assumption; their upper 10 cm contain on average 35% and 33% *N. pachyderma*, respectively. Although some down-slope process may account for the carbonate minima and red sediment on the Laurentian Fan, such processes cannot account for the evidence of warming during carbonate minima. If foraminifera were transported from shallower (northerly) locations, there would be a greater proportion of *N. pachyderma* during carbonate minima, suggesting cooling. Like most other North Atlantic locations, surface waters overlying MC-19 and -21 should register cooling during the LIA (*16*). Thus, we predict that the SST histories north and south of 44°N are out of phase on centennial to millennial time scales.

There are few other time series of the past

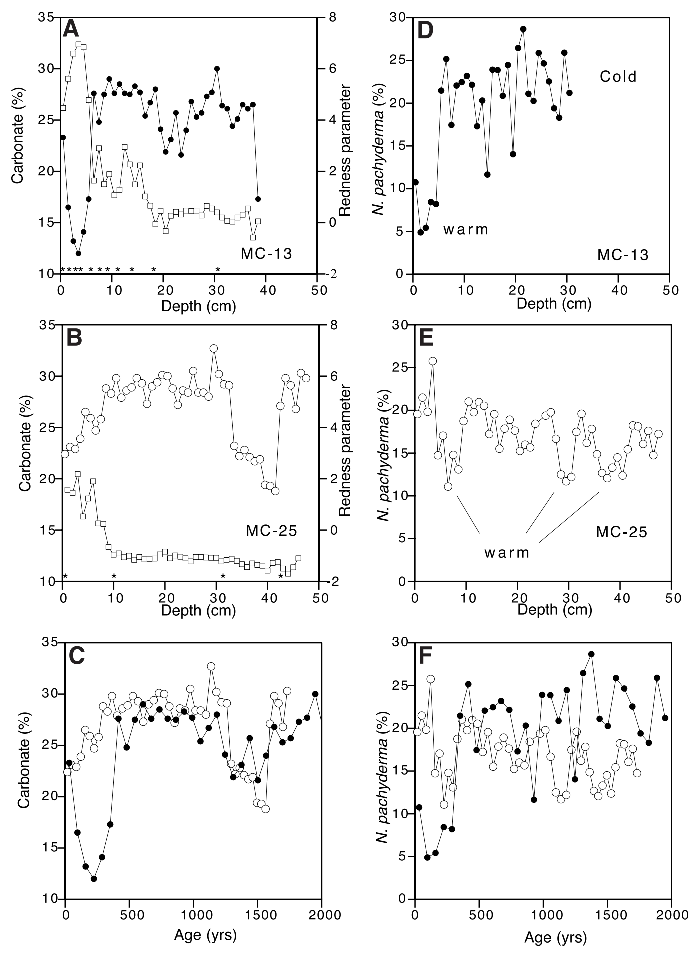

Fig. 2. Sedimentary and faunal data from MC-13 and -25. (**A** and **B**) Weight percent carbonate and "redness" (red:yellow color balance as shown by open squares) as a function of depth at MC-13 and -25, respectively. Carbonate data are solid circles in (A) and open circles in (B). Redder sediment has larger values of the redness parameter. Location of AMS ^{14}C dates (Table 1) are shown as asterisks along the depth axis. (**D** and **E**) Percentage of cold-water planktonic foraminiferal fauna (*N. pachyderma*) at MC-13 and -25 shown as solid and open circles, respectively. Each core has evidence of a warm event (minimum percent *N. pachyderma*) in the upper 10 cm and, as discussed in the text, higher deposition rate MC-25 resolves two older warm events. (**C** and **F**) Percent carbonate and percent *N. pachyderma*, respectively, plotted as a function of calendar age (B.P.) for MC-13 (solid circles) and MC-25 (open circles).

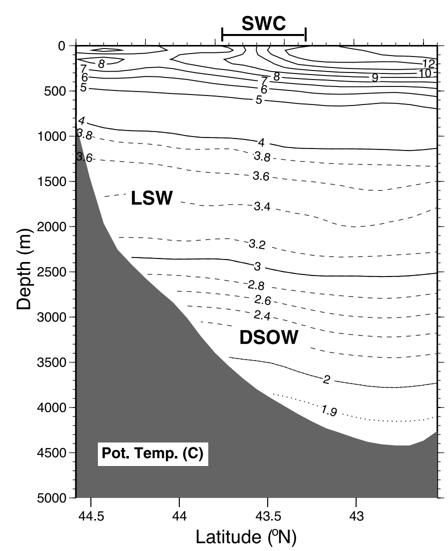

Fig. 3. Average slope water section of potential temperature (°C) for 55°W (*23*). Bar along the top denotes core of the slope water current as indicated by the compression of isotherms. Small shifts in the latitude of the slope water current (Fig. 1) could account for the SST changes we infer from planktonic foraminiferal faunal changes at our multicore sites. LSW = Labrador Sea water; DSOW = Denmark Strait overflow water.

2000 years to compare to MC-13 and MC-25. Results from the Bermuda Rise (*9*) are comparable in that the sedimentation rates are similar, bomb ^{14}C occurs at the sediment surface, and the cores can be correlated with percent carbonate (Fig. 4). At each location, carbonate minima are observed during the LIA and during the cold episode (1300 to 1500 years B.P.) that coincided with the Dark Ages. These are the youngest of a long series of millennial-scale climate events known from the North Atlantic (*9*, *17–19*). As discussed previously (*9, 17*), the coherence of these events may reflect some basinwide process capable of resuspending and advecting sediment, such as changes in the intensity of the deep recirculating gyres in the western North Atlantic. However, considering that our Laurentian Fan cores show some evidence of turbidity current transport and that the fan morphology in general is controlled by turbidity current erosion and overbank deposition (*6, 7*), this process must be important in delivering silts and clays to the Bermuda Rise via the Sohm Abyssal Plain (*20*).

The SST proxy of our Laurentian Fan cores is out of phase with SST on the Bermuda Rise (Fig. 4). Lowered SST over the Bermuda Rise during the LIA and the Dark Ages has been likened to the climatic response expected during a minimum phase of the NAO (*9*). Our evidence that the slope water current seems to have moved northward during the LIA, causing a local warming over the Laurentian Fan, is thus consistent with the notion that the slope water system oscillates on shorter (interannual to decadal) time scales in phase with the NAO (*2*). Hence, the marine geological data support the modern observations (where time series are relatively short and incomplete), and the physical oceanographic data help explain a surprising out-of-phase behavior in the SST proxy measurements.

Our data suggest that, if these marine sediments have been low-pass filtered by bioturbation, then either the NAO may have existed as a prolonged minimum state or minimum phase events may have been very extreme during climate oscillations like the LIA. However, analysis of Greenland ice core data does not support a strong NAO signal over the past 350 years (*21*). Another test of the NAO as a model for millennial-scale variability in the Holocene could come from paleo records of deep ocean circulation change. As mentioned earlier, available hydrographic data in the Laurentian Fan region suggest increased flow of Denmark Strait overflow water during the minimum phase of the NAO (*2*), but sediment grain size measurements imply decreased flux of Iceland-Scotland overflow water during the LIA and other late Holocene cold episodes (*19*). Thus, the proxy data and modern observations are contradictory in this regard, suggesting perhaps a different scenario for the deep ocean response on long time scales. If strong NAO-like variability in the surface ocean continued on century to millennial time scales, then the SST proxies on the Laurentian Fan should continue in antiphase with the Bermuda Rise and other North Atlantic locations through the Holocene and perhaps during other times before the last glacial maximum. During the peak of the last glacial maximum, however, there was probably little warm slope water present because the planktonic fauna over the Laurentian Fan was at least 50% polar, and subtropical fauna were absent (*22*).

Table 1. Accelerator radiocarbon dates on mixed planktonic foraminifera.

Depth (cm)	Accession number (OS)	Conventional ^{14}C age	Error $\pm$ 1 σ	Calendar age years B.P. (pre-1950)
		OCE 326 MC-13		
0.5	16709	> Modern*		0
1.5	16738	150	35	—
2.5	16739	420	55	—
3.5	16740	560	30	228
5.5	16741	720	30	328
7.5	16742	1200	30	720
9.5	17729	1370	30	913
11.5	16743	930	45	519
14.5	17731	1290	35	827
18.5	16744	1370	30	913
30.5	17047	2350	25	1948
		OCE 326 MC-25		
0	16746	> Modern*		0
10.5	17732	765	25	416
32.5	17730	1660	45	1221
43.5	17240	2030	25	1568

*Samples have a fraction modern (Fm) > 1, bomb ^{14}C is present. Fm = 1.0357 at MC-13 and 1.0087 at MC-25.

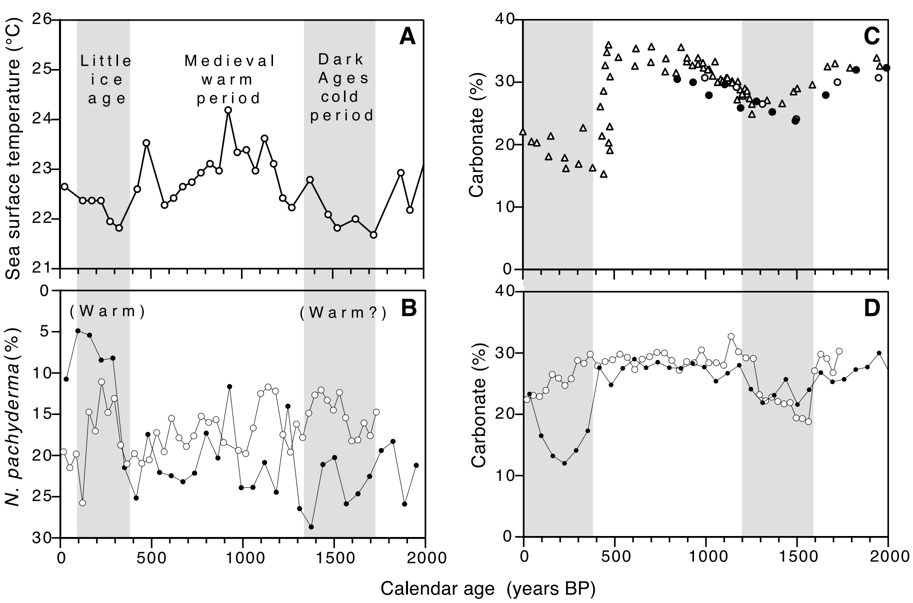

Fig. 4. Comparison of SST proxy data and sediment data from the northeastern Bermuda Rise in the Sargasso Sea (**Upper**) and the Laurentian Fan (**Lower**). Bermuda Rise SST estimates (*9*) (**A**) and Laurentian Fan faunal changes (**B**), which probably reflect SST changes of 1° to 2°C. In (B) both MC-13 (closed circles) and MC-25 (open circles) indicate warming during the LIA, which was a time of cooling in the Sargasso Sea, whereas warming during the Dark Ages cold period is supported only at MC-25. Similarity of carbonate records between the Bermuda Rise (*9*) (**C**) and the Laurentian Fan (**D**) [symbols as in (B)] requires widespread transport of noncarbonate clays and silts by deep recirculating gyres and by turbidites from the Laurentian Fan region to the Sohm Abyssal Plain and ultimately the Bermuda Rise (*17, 20*).

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variability of the deeper components of the slope water. Although variability of the deep components of this system is expected on NAO time scales (*3, 5*), there is no clear mechanism linking the NAO and the surface slope water current.
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8. Samples were extruded from the core tubes, sliced into 1-cm layers, and bagged. Subsamples were taken for micropaleontological study of the fraction >150 μm and for percent carbonate analysis with an automated gasometric technique. Sediment color was measured with a Colortron spectrophotometer and is presented using the "Lab" system [*CIE Colorimetry* **15.2** (1986)].
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10. As discussed in (*17*) and elsewhere [D. O. Suman and M. P. Bacon, *Deep-Sea Res.* **36**, 869 (1989)], Holocene carbonate cycles on the Bermuda Rise are driven mostly by pulses in the flux of terrigenous silts and clays.
11. The red clay-rich sediments that compose this facies are thought to derive from subglacial turbidite flow out of the Gulf of St. Lawrence [D. A. V. Stow, *Am. Assoc. Petrol. Geol.* **65**, 375 (1981)]. On the Scotian Margin, about half the glacial maximum to modern sedimentary section is this red sediment (*22*), and its thickness is even greater on the fan (*7*).
12. Accelerator mass spectrometer (AMS) ^{14}C measurements (Table 1) were made at the National Ocean Sciences AMS facility at Woods Hole on mixed planktonic foraminifera. Ages were converted to calendar years, according to Stuiver and Reimer [M. Stuiver and P. J. Reimer, *Radiocarbon* **35**, 215 (1993)], with a reservoir correction of 440 years. Age models were based on a linear fit of the ages to depth. For MC-25 the data are nearly linear, whereas for MC-13 there are age reversals of up to a few hundred years (Table 1). For this core, the age model is assumed to be linear from the origin to the deepest dated sample. Any other reasonable age model for MC-13 and MC-25 would still correctly identify the LIA near the core top, but ages of events deeper than about 10 cm in these cores are probably not known to better than ±200 years.
13. The relationship between percent *N. pachyderma* data from North Atlantic core tops and SST was presented by Kohfeld *et al.* (*15*). They showed that *N. pachyderma* blooms in the summer at high latitudes, whereas the only time series data on *N. pachyderma* growth in subpolar locations showed that the bloom occurs in the spring [L. R. Sautter and R. C. Thunell, *J. Foram. Res.* **19**, 253 (1989)].
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16. Although we do not have chronologies for these cores, the maximum percent *N. pachyderma* is found at the 3-cm level in each: 56% at MC-19 and 41% at MC-21.
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24. *Oceanus* cruise 326 and this research were funded by the National Science Foundation. We thank L. A. Conroy, K. Elder, C. E. Franks, J. Kelleher, E. Roosen, D. Torres, and the staff of the National Ocean Sciences AMS Facility for their assistance; R. Schmitt, I. N. McCave, and D. Oppo for reading the manuscript; and N. Driscoll, J. Grotzinger, and M. McCartney for helpful discussions.

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Multiple Ink Nanolithography: Toward a Multiple-Pen Nano-Plotter

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The formation of intricate nanostructures will require the ability to maintain surface registry during several patterning steps. A scanning probe method, dip-pen nanolithography (DPN), can be used to pattern monolayers of different organic molecules down to a 5-nanometer separation. An "overwriting" capability of DPN allows one nanostructure to be generated and the areas surrounding that nanostructure to be filled in with a second type of "ink."

Recently, there has been an intense effort to develop micro- and nanolithographic methods analogous to macroscopic writing and printing tools (*1–4*). These methods are allowing researchers to address important issues in biology (*5*) and molecule-based electronics (*6–9*). Microcontact printing (*2–4*) and even micropen writing (*1*) have been successful in terms of preparing molecule-based structures on the ~100-nm to centimeter length scale. We recently showed that dip-pen nanolithography (DPN) allows one to prepare custom, "single-ink" structures with dimensions on the sub–100 nm length scale (*10*). A significant issue in efforts to prepare nanolithographic printing tools pertains to registry—that is, how to use multiple inks within the context of one set of nanostructures spaced nanometers apart. At present, stamping procedures do not have the resolution capabilities of scanning probe lithographic methods or electron-beam (e-beam) methods, and with respect to multiple inks, they pose significant alignment problems (*4*). Moreover, traditional high-resolution techniques (*11–24*), such as electron and ion beam lithography and many scanning probe methods, rely on resist layers and the backfilling of etched areas with component molecules. These indirect patterning approaches can compromise the chemical purity of the structures generated and pose limitations on the types of materials and number of different materials that can be patterned. Indeed, adsorbate-adsorbate exchange can be problematic because a monolayer resist, which has surface binding functionality identical to that in the ink, is typically used in these methods (*18*).

We report the generation of multicomponent nanostructures by DPN (*10*) and show that chemically pristine patterns of multiple different materials can be generated with near-perfect alignment and 5-nm spatial separation. Additionally, we report an overwriting capability of DPN that allows one to generate a nanostructure and then fill in areas surrounding that nanostructure with a second type of ink. These demonstrations are analogous to the transition from (single ink) conventional printing to "four-color" printing, and should open many opportunities for those interested in studying molecule-based electronics, catalysis, and molecular diagnostics. Indeed, the spatial resolution of this multiple ink technique is similar to the length scale of conventional large organic molecules and many biomolecules (nucleic acids and proteins).

DPN relies on a water meniscus, which in air naturally forms between the tip and sample, as the ink transport medium, and therefore, one can use relative humidity as one method of control over ink transport rate, feature size, and linewidth (*10, 25, 26*). Before our invention of DPN, others attempted to develop scanning probe methods for depositing organic materials on solid substrates (*27*). They demonstrated deposition of micrometer-scale features composed of physisorbed multilayers of 1-octadecanethiol (ODT) on mica but concluded that under the conditions used ODT could not be transported to Au and, apparently, did not recognize the importance of humidity and the meniscus in the transport process. All DPN experiments were carried out with a ThermoMicrosopes CP AFM and conventional cantilevers (ThermoMicroscopes sharpened Microlever A, force constant = 0.05 N/m). To minimize piezo tube drift problems, we used a 100-μm scanner with closed loop scan control for all of the experiments. The ink in a DPN experiment can be loaded by using a solution method, which was described previously (*10*), or by using a vapor deposition

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