Method for characterizing self-assembled monolayers as antirelaxation wall coatings for alkali vapor cells

Y. W. Yi,[1] H. G. Robinson,[2] S. Knappe,[2] J. E. Maclennan,[1,a)] C. D. Jones,[1] C. Zhu,[1] N. A. Clark,[1] and J. Kitching[2,a)]

[1]*Department of Physics and Liquid Crystal Materials Research Center, University of Colorado, Boulder, Colorado 80309, USA*
[2]*Time and Frequency Division, National Institute of Standards and Technology (NIST), Boulder, Colorado 80305, USA*

We describe a method for characterizing self-assembled monolayers (SAMs) in terms of their performance as antirelaxation wall coatings for alkali atom vapor cells. A combination of initial surface analysis and subsequent laser spectroscopy is used to provide insight into the quality of the coating, as well as its performance under the exposure to alkalis as it occurs, for example, when used in applications such as atomic magnetometers or clocks. Fused silica plates coated with octadecyltrichlorosilane SAMs were used to make cubic ^{87}Rb gas cells. The surface was characterized by x-ray diffraction, contact angle measurements, and atomic force microscopy. Measurements of hyperfine resonance linewidths and frequency shifts show that the rubidium vapor atoms collide up to 40 times with the walls of the cells before their coherence relaxes and their adsorption energy is around 0.065 eV. Chemical analysis of the cell indicates some weak reactions between the coating and the rubidium atoms. © *2008 American Institute of Physics.* [DOI: 10.1063/1.2958329]

I. INTRODUCTION

Highly miniaturized instruments such as chip-scale atomic clocks (CSACs) and chip-scale atomic magnetometers (CSAMs) are being developed based on room-temperature atomic vapors confined in micromachined cells.[1–3] The precision of these devices depends on the relaxation time (T_2) of the appropriate atomic coherence; longer relaxation times lead to higher precision. The fast wall-induced depolarization of the atoms in evacuated vapor cells may be mitigated either through the addition of a buffer gas[4] to the cell or by applying an antirelaxation coating[5] on the interior surface. Buffer gases are usually inert gases or molecules that interact weakly with the alkali atoms. Frequent collisions of the alkali atoms with the buffer gas result in a slow diffusive motion, lengthening the relatively unperturbed observation time for atoms in the cell. However, buffer gases broaden the optical resonance signal that can degrade the stability of clocks and magnetometers,[6–8] and the presence of buffer gases can cause temperature-dependent shifts of the clock frequency.[9] In addition, the frequent collisions of the alkali atoms with the buffer gas interfere with the efficient creation of an alignment of the spins in the alkali atoms.[10] This makes it more difficult to use otherwise attractive spectroscopic techniques based on frequency-, amplitude-, or polarization-modulated light fields or optical-microwave double resonance that require atomic alignment.[11–13] These issues become particularly important in millimeter-scale instruments, where buffer-gas pressures approaching 100 kPa are needed to optimally suppress the wall-induced relaxation.

It has been predicted[14] that millimeter-scale atomic clocks based on wall-coated vapor cells could achieve stability comparable to those based on buffer-gas filled cells, if the walls allowed atoms to undergo a few hundred collisions before decoherence.

Wall coatings of alkanes with terminal methyl groups, which have single bonds terminated with hydrogen, can prevent spin relaxation and interact very weakly with alkali gas atoms.[15] A variety of wall coating materials based on long carbon chains have been studied for alkali metal cells. For example, films of paraffins such as polyethylene, Paraflint $[CH_3(CH_2)_nCH_3]$, and tetracontane $[CH_3(CH_2)_{38}CH_3]$ were used to achieve resonance linewidths of 1–10 Hz for rubidium gas atoms in a glass cell from several millimeters to several centimeters in diameter, corresponding to several thousand bounces before relaxation.[7,13,15–18] However, such so-called "wet films" are not compatible with current CSAC/CSAM fabrication methods and operation because of their relatively low melting points. Most microfabricated vapor cells are currently sealed using anodic bonding processes[19] that require temperatures above 250 °C.[20] At these temperatures, paraffin films tend to react with the rubidium vapor. It is therefore desirable to find alternative surface coatings that are compatible with cell fabrication techniques developed for chip-scale atomic devices.

Silane-terminated self-assembled monolayers (SAMs) (so-called "dry films") of alkyl chains chemically bond to glass and silicon oxide surfaces to form closely packed and ordered hydrocarbon layers of molecular thickness, as shown in Fig. 1.[21,22] Such coatings are thermally stable up to 230 °C in the air.[23] The hydrocarbon chains and the outermost chemical group (methyl) are essentially the same as in paraffins. They should therefore extend the coherent lifetime

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a)Authors to whom correspondence should be addressed. Electronic addresses: joseph.maclennan@colorado.edu and kitching@boulder.nist.gov.
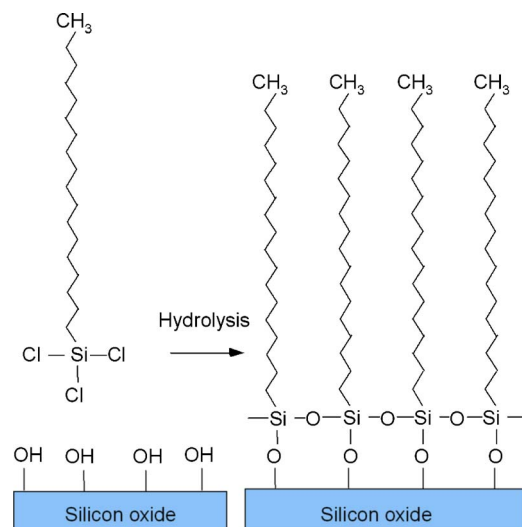
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FIG. 1. (Color online) Formation of OTS SAMs on a hydroxylated silicon oxide surface. The OTS molecules bond covalently to the substrate through hydrolysis and form a dense network. The carbon chains become oriented and extended by the van der Waals interactions between them during thermal annealing.

FIG. 2. (Color online) AFM images of fused quartz before and after coating with the OTS SAM. The images are 1 μm wide. The insets show water drops ($\phi \sim 1$ mm) on the surfaces. The OTS SAM is clearly much more hydrophobic than the bare quartz.

of excited atoms in Rb vapor cells. Camparo[24] studied Surfacil SAMs as antirelaxation wall coatings for the hyperfine coherence in Rb and reported two to three bounces of Rb atoms without depolarization. Stephens *et al.*[25] reported that octadecyltrichlorosilane (OTS) SAM-coated walls have lower cesium adsorption energy than bare Pyrex walls, but the difference in the energies was small. Surprisingly, all of these dry films showed much faster relaxation than paraffins even though the terminal groups were the same. Seltzer *et al.*[26] recently reported measuring 2100 collisions before the population relaxes (T_1) in a K vapor cell with a thick coating of OTS—although the relaxation times were not entirely reproducible. While it is well known that lighter alkali atoms show much less depolarization than heavier alkalis on colliding with cell walls,[27] this is nevertheless a very encouraging result. However, the quality of the SAM coatings was not well evaluated in the previous work; most techniques for evaluating surfaces are also difficult to apply for the round geometry of the cells. It is well known that the surface coverage of SAMs critically depends on the details of the preparation conditions.[28,29] We propose that knowledge of the structure and integrity of wall coatings can be very helpful in implementing them effectively and finding new types of wall coatings. In this paper, we describe cubic cells assembled from flat substrates coated with OTS SAMs of verifiably good quality that perform better than antirelaxation coatings in [87]Rb cells than any other SAMs previously studied.

The goal of the experiments described here was to identify wall coatings that could be reliably made and that resulted in the longest possible transverse spin-relaxation times. A method for characterizing such coatings in terms of their quality (e.g., surface coverage and thickness) and their interaction with alkali atoms (e.g., reactivity and relaxation times) was developed. The planar geometry of the cell allowed for easy surface analysis using x-ray diffraction, contact angle measurements, and atomic force microscopy
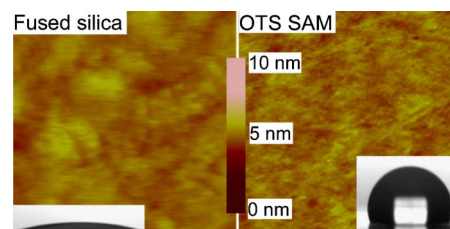
(AFM). From these measurements, it was possible to infer surface coverage, thickness, and roughness. The SAM-coated vapor cells were evaluated with respect to their ability to preserve atomic coherence by measuring hyperfine and Zeeman resonance linewidths, and the hyperfine resonance frequency shifted from the free space value. From these measurements, the adsorption energy and the number of coherent wall collisions were inferred. Finally, the cells were tested for reactions between the surface and the Rb atoms.

II. EXPERIMENTS AND RESULTS

A. Preparation of the coating

OTS SAMs were prepared according to a procedure described in the literature[30] with the addition of an annealing process. We chose this procedure because it is straightforward and includes a thorough cleaning process. First, rectangular quartz plates 0.5 mm thick (three 8×8 mm^2, two 8×9 mm^2, and two 9×9 mm^2) were cleaned in successive ultrasonic baths of acetone, isopropanol, distilled water, and de-ionized water for 5 min each. The plates were then dried in a stream of clean nitrogen gas and put in a "piranha" solution (60% sulfuric acid and 40% hydrogen peroxide by volume) at 90 °C for 30 min. Then they were rinsed with de-ionized water and dried again with nitrogen gas. The plates were immersed in an OTS solution (~ 1 mM in toluene) at ~ 20 °C for 30 min. The OTS solution was prepared in a nitrogen gas environment to avoid degradation by water in the air. The slides were then rinsed in turn with toluene, acetone, chloroform, and isopropanol. Finally, the SAMs were annealed on a hot metal block kept at 150 °C in a vacuum (3×10^{-2} kPa) overnight.

B. Surface analysis of the SAMs

The SAMs were characterized by AFM, x-ray reflectivity and contact angle measurements. The contact angle of a sessile water drop on an alkylsilane SAM is a convenient and traditional indicator of SAM quality; a larger angle indicates a more hydrophobic and hence closer-packed film. A circle was fit to a digital image of a de-ionized water drop (~ 1 μl) on the OTS SAM (see Fig. 2 inset) to obtain the contact angles. Measurements were performed with water drops at three or four locations on each SAM. The contact angle of the test OTS SAM was $110.00° \pm 0.4°$; the angle was increased to $111.8° \pm 0.5°$ after reannealing overnight.[31] The contact angle of the OTS SAM on a silicon wafer used
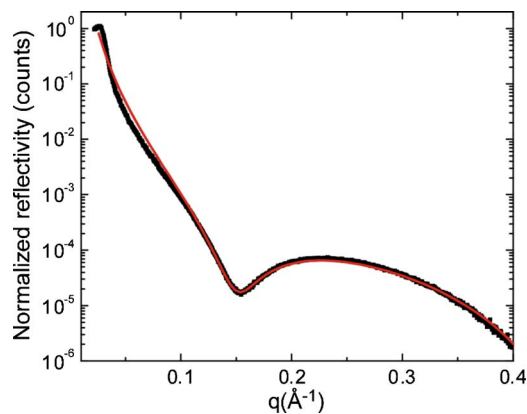
FIG. 3. (Color online) X-ray reflectivity of the OTS SAM on a silicon wafer. Measurements were performed with an 18 kW rotating anode x-ray source and a Huber four circle goniometer. The fit, obtained with a model assuming an alkane SAM with homogeneous electron density, corresponds to a wall coating with a thickness of 21 Å and surface roughnesses of 2.0 Å and 2.5 Å at the silicon and air interfaces, respectively.

for x-ray measurements was $111.8° \pm 0.4°$. These values are similar to those of OTS SAMs reported in the literature, which are between 109° and 114°.[21,22,32–34] The increase in the contact angle obtained by reannealing indicates incomplete packing of the initial SAM structure (see discussion below). We will show later that the annealing improves the antirelaxation properties of the SAMs. We also noticed small differences in SAMs made from different batches of OTS solution with freshly opened bottles of OTS, giving SAMs the biggest contact angles.

The effective surface coverage f_{OTS} can be calculated from the contact angle through the use of the following equation derived from Cassie's law,[35] assuming that the surface is composed of two components, a pure OTS SAM and a bare fused silica:[36]

$$f_{OTS} = (1 - \cos \theta)/(1 - \cos \theta_{max}). \qquad (1)$$

The angles θ and θ_{max} are, respectively, the measured contact angle of the sample and the maximum contact angle ever measured for the OTS SAM. We assume an ideal contact angle for bare fused silica of 0°. Taking $\theta = 111.8°$ and assuming that $\theta_{max} = 114°$,[21,34,36] we obtain an average coverage of $97.4\% \pm 0.6\%$ for our SAMs. The error mentioned here is just the statistical error from the measurements and does not include the uncertainty in the knowledge of θ_{max}, for example.

Contact mode AFM measurements (shown in Fig. 2) show little difference in the root mean square (rms) surface roughness of a bare quartz substrate (2.64 Å) and the same substrate coated with the OTS SAM (2.74 Å). The image of the OTS SAM surface is sharper than that of the bare quartz, presumably due to different interactions of the original and chemically modified surfaces with the AFM tip[37] rather than to actual topological changes in the quartz surface.

To further assess the quality of the coatings, we measured the x-ray reflectivity of SAMs prepared on silicon wafers. The model used to fit the reflectivity data plotted in Fig. 3 is of a silicon substrate that is covered uniformly with an organic layer that is taken to be uniform in electron density
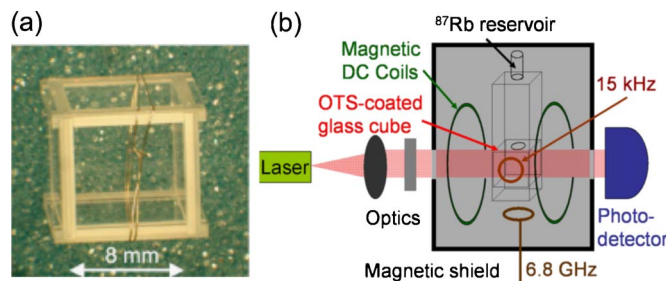

FIG. 4. (Color online) (a) Photograph of an assembled fused silica cube. (b) Schematic of the cube in the spectroscopic setup. A DBR laser optically interrogates the ^{87}Rb atoms in the cubic cell. Mounted below the cell is a single-turn coil oriented to excite the $\Delta F = 1$ and $\Delta m_F = 0$ microwave hyperfine transition. Mounted on the side of the cell is a five-turn coil ($\phi \sim 1$ cm) oriented to excite the $\Delta F = 0$ Zeeman transition. The absorption of the light was measured as a function of the excitation frequency.

(the "one box" model[38]). This model yielded an OTS SAM thickness of 21 Å, with the roughness of the silicon-OTS and OTS-air interface found to be 2.0 and 2.5 Å, respectively. This thickness is about 10% smaller than the highest previously reported values of 23 and 23.6 Å.[21,38] This result implies that the areal packing density (number of molecules/ unit area) is about 10% lower than the best reported OTS SAMs. The molecular tilt (the average angle between the hydrocarbon chains and the surface normal) of the SAM on silicon is calculated to be 34°, slightly larger than the 20° tilt reported in the literature.[38] Even though the films are thinner than usual, the measured molecular coverage is nevertheless very good.

C. Preparation of the vapor cell

Six individually coated fused silica slides were assembled into a closed container with cubic geometry. By coating the planar surfaces prior to assembly, the inside walls of the cube could be uniformly and reproducibly coated, and tested for quality with the evaluation processes described above.

In a typical experiment, seven fused silica slides were prepared in the way described above. Six of these were assembled into a cube [like the one shown in Fig. 4(a)], and one was kept for surface analysis. The cells had grooves cut into them for easier assembly and were held together with several turns of nonmagnetic wire, completely avoiding adhesives that could have potentially contaminated the surfaces at elevated temperatures or have reacted with the rubidium.

The cube was then inserted into a glass cell in the form of a rectangular parallelepiped 4 cm long and with a square cross section of 12×12 mm^2. A stem was attached to one end. In addition to serving as a Rb reservoir, the stem allowed evacuation of the cell before being filled with ^{87}Rb. A small break-seal connection to a secondary open tube allowed for reattachment to a vacuum system after the cell assembly was pulled off. This port would later be used for re-evacuation and background gas evaluation. This cell assembly was then attached to a glass vacuum manifold along with a second glass break-seal containing isotopically enriched ^{87}Rb. The cell was evacuated and the oven temperature gradually increased to ~ 200 °C for over 6 h. The total
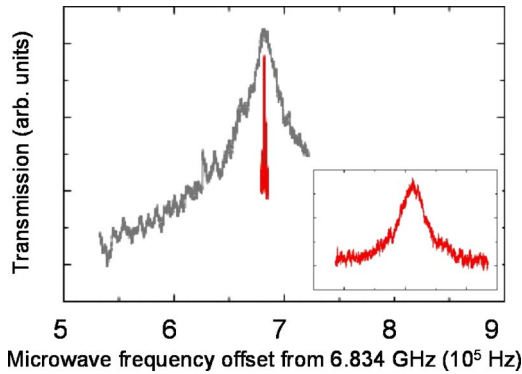
FIG. 5. (Color online) Transmission spectrum of the 0–0 hyperfine micro-wave resonance as the microwave frequency is scanned in the OTS cube (red) and in an uncoated cell of similar geometry (gray). The inset is a close-up view of the OTS lineshape, showing a linewidth of a few kilohertz.

bakeout time was ∼22 h, after which the pressure dropped to ∼3×10^{-6} kPa at 200 °C. Finally, after the Rb break-seal was opened and the Rb distilled into the reservoir section, the cell assembly was pulled off. Two cells were made according to this procedure.

D. Spectroscopy

The cell assembly was mounted inside an oven designed so that the temperature of the cell walls and the rubidium reservoir could be controlled separately. The residual magnetic field produced by a current flowing through the oven heater was estimated to be less than 1.5 nT. The current was ac modulated, and measurements were taken only when the current was off. A distributed Bragg reflector (DBR) laser tuned to the D_1 line of Rb was used to optically interrogate the [87]Rb atoms in the atomic vapor. Mounted below the cell was a single-turn coil ($\phi \sim 0.5$ cm) oriented to excite the $\Delta F = 1$ and $\Delta m_F = 0$ microwave hyperfine transitions at 6.8 GHz. Mounted on the side of the cell was a five-turn coil ($\phi \sim 1$ cm) oriented to excite the $\Delta F = 0$ Zeeman transition at 15 kHz [see Fig. 4(b)].

For measurement of the hyperfine resonance, linearly polarized laser light was used, which was tuned to the $F \rightarrow F' = 2 \rightarrow 2'$ transition. A ∼2.5 T magnetic field along the direction of the laser beam splits the degeneracy of the Zeeman levels. When a ∼6.8 GHz microwave signal was applied to the loop, the light absorption increased as the microwave frequency approached the splitting between the $m_F = 0$ levels. The width and frequency of the resonance between the $m_F = 0$ levels were measured with respect to a hydrogen maser reference at low light intensity and microwave power. The resonance frequency was measured by lock-in detection of the microwave source, with its frequency modulated at a 10 Hz rate. The linewidth was obtained from a Lorentzian fit to data obtained by amplitude modulation of the applied microwave (or Zeeman) power. Typical transmission spectra are shown in Fig. 5 for one of the OTS-coated cubes (red spectrum) and an uncoated cell of similar geometry (gray spectrum).

The measured width of the hyperfine resonance was in the range of 800–1100 Hz, depending on the cell temperature (see below), while the resonance was shifted from that of the

unperturbed atom (6.834 GHz) by about −600 Hz. Both parameters are used to characterize the effectiveness of wall coatings in preserving hyperfine coherence. We found that the linewidth decreased over the duration of hours, indicating some "curing" of the walls.

The average number of bounces undergone by a typical atom before its hyperfine coherence relaxes is given by the ratio of the coherence time T_2 (derived from the rf resonance linewidth) to the time between wall collisions τ_c,

$$N_B = \frac{T_2}{\tau_c}, \tag{2}$$

where $\tau_c = \langle d \rangle / v_{\mathrm{Rb}}$ is given by the average distance d between the wall collisions ($d = 5.3$ mm) and the Rb velocity v_{Rb}. The measured linewidths correspond to ∼30 bounces at 170 °C and ∼21 bounces at 102 °C. We estimate the contributions of spin-exchange collisions and power broadening of the linewidth to be below 50 Hz. A surface coverage of less than 100% could contribute to a large fraction of this linewidth (see discussion).

In the case of Zeeman pumping, circularly polarized light was used. The laser was resonant with the $F = 2$ $F' = 1$ transition, and a small longitudinal magnetic field of ∼2.5 T was applied. The width of the Zeeman resonance was measured at low light intensity and rf power. The Zeeman linewidths were 562 Hz at 82 °C and 444 Hz at 102 °C, giving a ratio of 0–0 hyperfine structure width to Zeeman width of only about 1.6 and corresponding to about 31 and 41 coherent bounces, respectively. This is a somewhat surprising result since previous measurements of [87]Rb on alkane coatings showed that the Zeeman linewidth can be narrower than the hyperfine linewidth by as much as a factor of 10.[16] Furthermore, when the cell walls were kept at the same temperature as the Rb reservoir, the wall quality slowly degraded over time. We attribute this degradation to Rb sticking to the walls; the initial linewidths can be recovered by heating the walls far above the Rb reservoir temperatures. We conclude that control of thermal gradients in the cells is more important in coated cells than in cells filled with buffer gas.

To our knowledge, the results for the OTS reported here display the highest number of coherent collisions for the 0–0 hyperfine transition measured for [87]Rb atoms with any SAM surface. At the same time, it is an order of magnitude worse than the results from cells with (thicker) paraffin and OTS coatings.[15–18,26] OTS SAMs could nevertheless be a useful candidate for antirelaxation wall coatings in applications where higher temperatures (above 100 °C) are needed and buffer gases are problematic.

E. Chemical stability

In order to monitor the occurrence of any chemical reactions between the rubidium and the walls, the cell was re-evacuated after a period of initial testing by use of a glass break seal attached to the body of the cell. The gas compounds were analyzed with a mass spectrometer. Figure 6 shows the mass spectral data for the background gas inside one OTS cell, indicating that some rubidium did react with the walls. Fragments of the hydrocarbon chains are present,
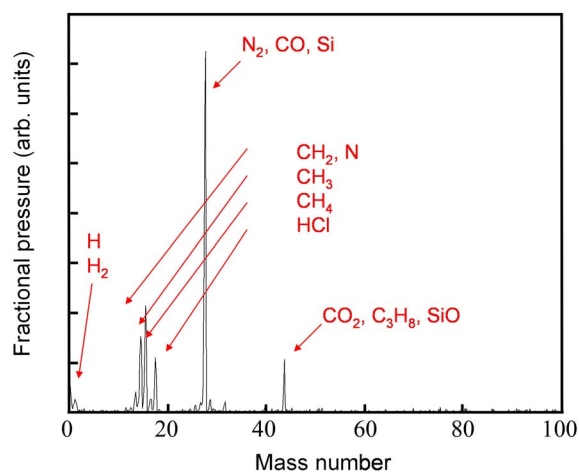
FIG. 6. (Color online) Mass spectrum of the cell background gas after exposure to Rb for several days. Some possible molecular fragments are identified.
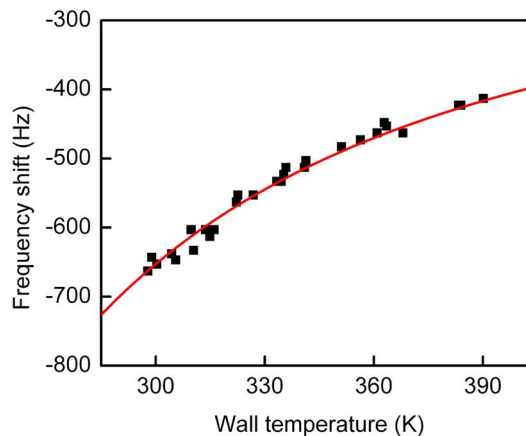

FIG. 7. (Color online) Shift of the 0–0 hyperfine resonance from its unperturbed value, measured as a function of the cell wall temperature (black squares). The line is a fit using Eq. (3).

such as methane (CH_4), methylene (CH_2), and methyl (CH_3), similar to the fragments found by Camparo *et al.*[39] in cells with Surfacil coatings after exposure to Rb. A major difference in the spectra is the presence of a peak at a mass of 44 in the OTS cells, while Camparo *et al.*[39] measured significant fractions at masses of 43 and 58 that are missing here. Even though a gas pressure built up inside the cell, the resonance linewidths after re-evacuation were found to be similar to the values measured prior to evacuation.

While these subtle reactions between the alkali atoms with the OTS surface do not seem to harm the performance of the cells at lower wall temperatures, irreversible damage to the walls resulted when the cell was heated to temperatures around 190 °C. Without the presence of rubidium, however, the coatings are thermally stable up to 230 °C.[23]

F. Adsorption energy

Adsorption energies have been measured for a number of alkalis and surfaces.[15,24,40,41] Since the adsorption energy E_a is related to the period the alkali atoms spend physically adsorbed on the wall of the cell, which in turn largely determines the wall-induced relaxation rate, this quantity can give some insight into the effectiveness of antirelaxation coatings. E_a can be deduced from $\delta\nu$, the resonance frequency shift of the 0–0 hyperfine resonance, when measured as a function of the absolute wall temperature T,[40]

$$|\delta\nu| \propto \sqrt{T}e^{E_a/kT}. \tag{3}$$

The dependence of the frequency shift on the wall temperature in an OTS cell is shown in Fig. 7. The temperature of the Rb reservoir was kept slightly lower than the temperature of the cell wall to prevent deposition of Rb onto the wall coating. A fit to the data returned a value of $E_a = 0.065 \pm 0.002$ eV for the adsorption energy. This is nearly an order of magnitude lower than the values reported for OTS and cesium,[25] and for (poly)dimethylsilozane and rubidium.[42]

It can be seen from Fig. 7 that the magnitude of the frequency shift decreases with increasing wall temperature.

At the same time, the linewidth of the hyperfine resonance decreases from 1.07 kHz at 50 °C to 870 Hz at 102 °C. Finally, we note that the cell was operated for extended periods at temperatures above 170 °C, with no degradation of the performance.

III. DISCUSSION

Although the spin-relaxation rate of Rb gas cells coated with OTS SAMs is an order of magnitude smaller than that of the coatings with Surfacil SAMs, it is still an order of magnitude larger than conventional paraffin coatings. Since, apart from the silane head group, the chemical structure of OTS SAMs is the same as paraffin, the difference in effectiveness in preventing spin relaxation may well be due to an incomplete coverage, a lower molecular packing fraction of the SAMs, and the difference in their thickness. A coverage of only 97.4%, as estimated from the surface measurements, would result in a linewidth of 420 Hz for the given cell geometry. This is very close to the Zeeman linewidth measured in the experiment. It might be the case that the majority of this linewidth is due to the imperfect coverage of the SAMs rather than the quality of the actual coating. This might also explain the fact that Seltzer *et al.*[26] measured much narrower lines in thick OTS coatings, as multilayers could more likely result in better overall coverage. In order to reach 1000 bounces in a SAM-coated cell, the coverage is required to be 99.9%, which seems challenging with the current SAM coating methods.

Furthermore, there are a number of uncertainties about the remaining coatings even after taking into account the results from the surface analysis; the SAM surfaces could have been contaminated during the assembly of the cube. For example, a cell fabrication process that is established to work well for wet films might not yield good results for SAMs because of the absence of a "self-curing" process, such as remelting of the coating after assembly of the cell. Finally, we have not taken into account the fact that the edges and corners of the cubes are not completely sealed. Rubidium

atoms could get trapped in these gaps or leave the cell, which would decrease the coherence lifetime and therefore the effective number of bounces.

Even if the coverage of the Surfacil SAMs is close to unity, as argued by Camparo,[24] they are much thinner than OTS SAMs covering the substrates only with methyl (CH_3) groups while OTS SAMs comprise longer hydrocarbon chains [$(CH2)_{17}CH_3$]. Further investigation of relaxation dynamics using more densely packed OTS SAMs and silane-based SAMs with different chain lengths would help us to better understand the relaxation mechanism of Rb coherence on SAMs. Coating different substrate materials with the same coating could furthermore give insight into the effect of surface roughness on the atomic coherence as well as the possibility that the alkali atoms might "see" substrate properties, e.g., magnetic impurities, through the thin coating film. Finally, a comparison between population (T_1) and coherence (T_2) relaxation times could allow for further understanding of the relaxation mechanisms. While the interaction of such coatings with alkali atoms is still not completely understood, experimental results of the combination of methods described in this paper might be a step to foster better comprehension. We hope that it will result in our ability to choose coatings based on their chemical and physical properties, and predict their performance as antirelaxation wall coatings for alkali atoms.

IV. SUMMARY

We have investigated the performance of OTS SAMs as antirelaxation coatings for atomic vapor cells prepared from flat fused silica plates to allow prior characterization of the SAM surfaces. Contact angle measurements suggest an average coverage of $97.4\% \pm 0.6\%$ for our SAMs. X-ray diffraction analysis gives the OTS SAM a thickness of 21 Å, with the roughness of the silicon-OTS and OTS-air interface found to be 2.0 and 2.5 Å, respectively. This result implies that the molecular packing density is lower than the best reported OTS SAMs by about 10% and the tilt angle of the alkyl chains is 34°. Contact mode AFM measurements show little difference in the rms surface roughness of a bare quartz substrate and that of the same substrate coated with the OTS SAM.

Rubidium vapor atoms were measured to collide up to 40 times with the walls of small cubic cells before their coherence is lost. The cells have been successfully operated at temperatures up to 170 °C without loss of performance despite some chemical changes in the coating after exposure to the rubidium vapor. The overall stability of silane-based SAMs in the performance of antirelaxation at high operating temperatures makes these materials promising candidates for antirelaxation wall coatings in alkali vapor cells. The effective coverage and the packing density are close to those of previously reported OTS SAMs of high quality. Since this coverage is estimated to result in resonance linewidths very close to the Zeeman linewidths measured in our experiments, we infer that the minor imperfections in the SAM limit the number of collisions occurring before decoherence. Finally,

the adsorption energy of the coated walls was measured to be nearly an order of magnitude lower than that previously reported for silane-based SAMs.

We propose that systematic and independent physical characterizations of wall coatings will be a useful tool for developing coatings satisfying the rigorous requirements of CSACs and CSAMs. The combination of surface analysis with spectroscopic measurements can help in understanding which properties are important in creating antirelaxation coatings of good quality. In order to achieve significantly higher numbers of coherent collisions in alkali gas cells, further improvement in the OTS SAM quality seems necessary. In addition, experiments with SAM molecules with different lengths and terminal groups, as well as theoretical study of the interaction of alkali atoms with wall coating materials, should be interesting.

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