Re: AJ11387

Measurements of the ground-state polarizabilities of Cs, Rb, and K using atom interferometry
by Maxwell D. Gregoire, Ivan Hromada, William F. Holmgren, et al.

This document is our reply to the referee's comments.

We thank the referee for providing helpful comments on our manuscript. We also received helpful feedback from three other colleagues. Guided by these comments and suggestions, we have improved our manuscript. The manuscript is now ready to be resubmitted for publication in Physical Review A. Below, we reply to each comment made by the referee. Then we provide a summary of additional changes that we have made.

Referee comment:

"1) Sect. 2C, 1st paragraph, line 13: it is stated the pillars (which produce the electric field) are moved 400 microns in order for the atomic beam trajectory to shift from one edge of the gap to the other. Earlier in this paragraph, it is stated that the gap is approximately 4000 microns. Is there a typo here? If not, this apparent discrepancy should be explained."

Our response:

To make this part of the paper (Section II.C. Paragraph 1) more clear, we revised the sentences in question to say: "We begin a polarizability measurement with the assembly positioned such that the beam passes through the gap between the pillars near one of the edges. We take 25 sec of data with the electric field on and 25 sec with it off. We then move the pillars in nine 400 μm increments so that the beam approaches the other edge of the gap, taking 50 sec of data at each location."

There was not a typo nor a discrepancy, but perhaps the earlier description was confusing. So in response to the referee's comment we revised the description of our experiment in an effort to avoid confusion. To say it one more way, the gap width is approximately 4000 um. We measure the electric field induced phase shift of the interference pattern with the electrodes located in 10 different positions with about 400 um between each position. This is how we acquire data shown in Figure 6.

Referee comment:

2) Fig. 7: The grating period d_g is given as 100.0(1) nm. How is this uncertainty determined? Since this is one of the dominant contributions to the overall uncertainly, this should be described somewhere in the text (or a reference given).

Our response:

We thank the referee for raising this question. We asked Tim Savas, the manufacterer of the gratings, for more information about the grating period and period uncertainty. From his response we learned that we should use a value for the period of 99.90(5) nm. This is different (but a consistent with) the period of 100.0(1) nm that we used in our earlier analysis. Now in the revised manuscript we adjusted our analysis to use this new value for the period.

To share with the referee what we have learned, we quote verbatim part of our email exchange on this topic.

---------- Forwarded message ----------
From: **Tim Savas** <tim.savas@gmail.com>
Date: Thursday, October 15, 2015
Subject: nanogratings
To: Maxwell Gregoire <maxwellgregoire@gmail.com>
Cc: alex cronin <cronin1010@gmail.com>

Hi Maxwell,

Your groups' question slipped my mind. Sorry. I spent this morning searching files on my computer, searching old emails, and searching files in my brain. The latter was the most difficult -- it's been a while since this question has arisen.

My email only goes back to 2006, I must have lost the old files when transferring stuff to a newer laptop. I did find a few places where I told people p = 99.90 +- 0.05 nm, and told them it was a measurement performed by NIST. (More about this later.)

I also thought Peter Toennies' group at one of the MPIs in Germany independently measured the pitch, but I can not find any info on that. I did see a paper where they used the 99.9 +- 0.05 value. This morning I emailed Wieland Schoellkopf (a grad student in Toennies' group at the time) asking him if they did a measurement, and if so, what are the details? I haven't received a reply yet.

The NIST-measured sample was done by another customer and communicated to me by email. I no longer have that email, but what I remember is the customer (with whom I have an NDA, which is why I'm being a little secretive even though I don't like it) said NIST used their C-AFM to do the measurement. Perhaps if you do a google search of NIST and C-AFM you can find some info on the way they do the measurement and treat the uncertainties. There is no way my customer is going to share detailed info on the measurement.

At some point somebody should do a measurement! I'll let you know what Wieland says, assuming he responds. Maybe you guys can send a chip (with broken membranes?) to NIST, or maybe we can think of a good way to do a measurement.

So, a sentence you can use in the meantime (feel free to edit as you wish):

"A grating in silicon, produced by the same achromatic interference tool used to produce the free-standing nitride gratings, was sent to NIST and measured using their C-AFM tool."

Okay, that sentence will need some work, I think. The point is the achromatic interference lithography tool produces the same pitch for every (daughter) exposure. The pitch of the daughter is determined by the pitch of the parent gratings which is fixed.

I hope the info helps, and I'm sorry I can't give a better answer. I also hope Wieland replies.

Regards,
Tim

> On Oct 14, 2015, at 4:41 PM, Maxwell Gregoire <maxwellgregoire@gmail.com> wrote:
>
> Hi Tim,

Have you found any more information yet on why the nano gratings have 0.1% uncertainty in period? Even if you can't find an exact number for the uncertainty, would you be able to give me a sentence or two on why that uncertainty is what it is? I'd like to put a sentence about it in the PRA (as per referee comments).

Thanks for your help,
Maxwell

After asking some follow-up questions, Savas responded with more information regarding the uncertainty:

---------- Forwarded message ----------
From: **Tim Savas** <tim.savas@gmail.com>
Date: Thu, Oct 15, 2015 at 3:46 PM
Subject: Re: nanogratings
To: Maxwell Gregoire <maxwellgregoire@gmail.com>
Cc: alex cronin <cronin1010@gmail.com>

Maxwell,

Yes. The assumption is every daughter grating has the same period; however, there are some details.

Let's first define terminology so we don't have confusion. Sometimes "lithography" just means the initial patterning part (the exposure, in this case) and sometimes it refers to the whole process. For our discussion, let's separate the fabrication into two parts: the exposure and the processing. Let's discuss the exposure first.

There is some thermal expansion in the parent gratings and mounting hardware, and I never tried to thermally stabilize the tool. Let's say the lab temp didn't vary more than a few degrees C. In that case, I think thermal expansion effects are no larger than ~ 100 parts per million for exposures done at different times -- from day to day.

Next, the alignment, from run to run, of the parent gratings and daughter-grating substrate also produces changes in period of the same order (or smaller). So, that is also under a part per thousand.

Finally, as far as the exposure is concerned, there is some chirp in the period of the parent gratings. That chirp is very small and can result in a pitch difference, from one chip to the next (over a large exposure area), of 10 or 100 parts per million.

Therefore, as far as the exposure is concerned, the period of every daughter grating is identical to within a part per thousand.

As far as the remaining processing, nothing can change the period. The initial pattern just gets etched down into the substrate. What can change the pitch however, is stress or thermal expansion in the membrane. I have not analyzed that. The membrane can buckle out of plane, for example, and that would change the pitch.

To wrap it up, yes, by measuring the period and uncertainty of one of the daughter gratings (on a solid substrate) at some temp, or one of the parent gratings, then the period and uncertainty of all the daughters at close to the same temp are known to a part per thousand.

We decided to revise our paper to use the period and uncertainty of 99.90(5) nm stated by Savas. This 0.1% change in grating period reduced our measured polarizabilities by 0.1%, which was a slightly smaller adjustment than the uncertainties in those polarizabilities. The decreased uncertainty in the grating period reduced the fractional uncertainties in our polarizabilities from around 0.18% to around 0.16%. We revised all values in the paper that were derived from those polarizabilities. Since the change in our polarizabilities was insignificant, our measurements still agree or don't agree with everything that they did or didn't agree with before.

The updated knowledge about the grating period did not affect the ratios of polarizabilities that we report.

We chose not to discuss how grating period and its uncertainty were determined in our manuscript because we don't have any first-hand experience in determining that uncertainty—we are simply using the manufacturer's stated value. In section II, paragraph 1, we now cite an article and a PhD thesis by Tim Savas:

Tim Savas et. al, "Achromatic interferometric lithography for 100-nm-period gratings and grids", J. Vac. Sci. Technol. B 13, 2732 (1995)

Tim Savas, PhD thesis from MIT, "Achromatic Interference Lithography."

Referee comment:

3) P. 10, col. 1, line 6: "Trubko2015 [17, 48, 75]" is confusing. Should this be "[30, 17, 48, 75]"?

Our response:

We have revised this and no longer refer to "Trubko 2015".   Instead, in Section III.C. we state that values of R (ratios of line strengths) are in Table V. Then in Table V we refer to references [72,65,79] in the revised manuscript.

The name "Trubko2015" was used to refer to a paper by our team that has not yet been submitted for publication. This work reports a new value for R (the ratio of line strengths) for potassium that is more precise, but consistent with published values (for example reference [79]).

The Editor commented:

Figure 4(b), 5, 7, 8, 9: For figures that are color online only, please ensure that the figures, captions, and text references to these figures are appropriate for both the online color and the print grayscale versions. Note that the same figure file is used for both versions. Many standard colors have similar grayscale values and therefore may be indistinguishable in the print version. For example, reference to a "blue line" and a "red line" in a figure is insufficient, as these labels are meaningless for the grayscale version. Mention shading or location as an additional description in the caption ["the green (light gray) line," "the green (upper) line"], or use different styles of curves ("dotted blue line," "dashed red line") or data point symbols ("blue circle," "red square") in the figures. (See http://journals.aps.org/ for more information.)

Our response:

We changed Figs 4(b), 5, 7, 8, and 9 accordingly. The three lines in 4(b) are now solid, small dashes, and large dashes. Figs 5 and 7 were converted to grayscale with black, gray, and white bars. Figs 8 and 9 now use markers with unique shapes for each color (rather than all the same shape regardless of color).

Editor comment:

Figure 6, 7, 8, 9 labels with subscripts: The lettering in the axis labels and/or numbering size should be increased.

Our response:

We increased the font sizes for labels with subscripts in Figs 6, 7, 8, and 9 accordingly.

Additional Changes:

We corrected the following errors and provided the following clarifications:

- Added “Ami02” to 6th entry in bottom-left graph in Fig 9 and cited it in the caption. Reference Bou07 presented a revision of the result in the Ami02.
- Error bars in 2nd entry in top-left graph in Fig 9 were calculated incorrectly. Fixed it.
- The result in reference Mit03 (in Fig 8 and Table 8) is actually semi-empirical, not ab initio. Corrected this in Fig 8, the Fig 8 caption, and Table 8.
- In Table 8, we indicated the method used in different ab initio calculations by adding acronyms to the “Method” column. These acronyms are consistent with previous publications (see J. Mitroy, M. S. Safronova, and C. W. Clark, J. Phys. B 44, 202001 (2010)) and are defined in the Table 8 caption.
- Removed the text “+Side” in Fig 1 because it was irrelevant to the paper (it referred to an older convention of defining the different interferometers that we did not use in this paper).
- Removed “dv” in Eq 5
- Wrote Eq 10 in terms of Einstein A coefficients instead of lifetimes, changed the sentence before Eq 10 accordingly.
- In section IIB, paragraph 5, sentence 2, the “v_0 = 1400 m/s” was a typo and should have been “v_0 = 2100 m/s”. The calculation (2100 m/s) / 14 = 150 m/s is now correct (and all the numbers in it are correct).
- Sec IIB, paragraph 10: added “See [29] for an explanation of how we measured θ_g.”
- Sec IIB, paragraph 11, sentence 3: changed “over time we measure” to “over time, typically 3% over the course of several hours, we measure”, changed “about” to “twice”
- Sec IIIA, paragraph 1, sentence 3: changed “4 times smaller” to “3 times smaller”
- Sec IIIA, paragraph 3, sentence 4: cited Derevianko et al (1999), ended sentence with “, and Derevianko and Porsev's later $\acs$ prediction \cite{Derevianko2001} was made using a

measured van der Waals C_6 coefficient."

We also made the following grammatical corrections and changes of word choice/phrasing:

- Sec I, paragraph 1, sentence 3: changed "many" to "several"
- Sec I, paragraph 4, sentence 2: changed "coupling strength, E_PNC, of" to "PNC amplitude E_PNC due to", removed "the" in "the electric dipole"
- Sec I, paragraph 5, sentence 3: changed "that" to "was"
- Sec I, paragraph 5, sentence 6: changed "detector obtain" to "detector to obtain"
- Sec IIA, paragraph 2, sentence 2: changed "1 eV in our" to "1 eV for Cs in our"
- Sec IIC, paragraph 6, sentence 1: changed "a_pillars to 0.5 um accuracy" to "the gap between the pillars, 2a_pillars, to 1 um accuracy" (The meaning is the same, the latter is stated more precisely.)
- Sec IID, paragraph 2, sentence 2: changed "obtain" to "estimate"
- Table IV caption, sentence 2: changed first and third instances of the word "errors" to "uncertainties"
- Sec III, paragraph 2, last sentence: changed "the Outlook section (Section IV)" to "Section IV"
- Sec IIIA, paragraph 3, sentence 3: changed "match our" to "match all three of our", cited Derevianko et al (1999)
- Sec IIIB, paragraph 1, sentence 2: changed "; those" to ". Those"
- Sec IIIC, paragraph 3, sentence 1: changed "coupling strength" to "amplitude"
- Sec IIIC, paragraph 3, sentence 2: added "as a test of the standard model" at end of sentence
- Sec IV, paragraph 1, sentence 1: changed "easily" to "accurately"
- Sec IV, paragraph 1, sentence 2: removed "direct measurement of"