

To the Editor,  
Scipost Physics.

We thank you for giving us the opportunity to address the reviewers' recommendations and resubmit the manuscript with minor revision.

We have tried to respond to all the recommendations made. The answers to the referees' questions and comments are given below. We have mentioned the text where ever we have made changes in the revised manuscript.

Furthermore, we find some typographical errors in our submitted version. We have corrected them in the revised manuscript. We also rerun our code to calculate the TDMS for the Stokes transitions, with the updated values of some of the constants given in Table 2 according to ref 59, and we provide the updated numbers in the revised manuscript.

We hope that after going through the answers, you will not hesitate to accept our manuscript for publication in your esteemed journal.

Thanking you.  
Kind regards,  
Arpita Das

### **Answers to the reviewer 1**

We thank the reviewer for his/her review of our manuscript by recommending it for publication with more consideration. He/she further posed some valuable questions. We have tried to address all the queries.

Q1. This paper reports on a very detailed and elaborated theoretical model to fully characterize the transition strengths of the transitions which would be involved in a STIRAP scheme to create ultracold RbCs molecules. In the way the paper is written, the reader understands that the model has been set up, and has been probed experimentally with the guide of the model. Without minimizing the quality and the usefulness of the model, I wonder if the experimentalists could have "simply" searched for the right transitions, starting from a simple model (namely, without the hyperfine structure) to locate the proper Franck-Condon zone, and then explore one by one the lines composing the hyperfine structure of the excited state, to identify the most suitable one. Could the authors comment a bit in this matter? My reasoning is simple: would the authors recommend to pursue such a theoretical modeling in any case prior to perform the experiment? If yes, I guess that such a "savoir-faire" should be proposed to the community in a way or another (lecture course, code sources)

A1. We agree that it is possible to search for the proper transitions using the experiment, and indeed, this is what the Innsbruck group has done previously to identify the best transitions at 181.5G. However, such a search can take a very long time to complete due to the large number of transitions available between the different hyperfine components in the excited state and the possibility of performing STIRAP from different Feshbach states. We found that this approach of having a model including the hyperfine structure significantly increased the efficiency of the search and so is useful even if not strictly necessary.

We have proposed a change to the text to explain how the previous work was done:

At 181.5 G, the transitions for STIRAP were found starting from a model without hyperfine structure, and so required an exhaustive search through the many available transitions by experiment [42]. Here we identify suitable transitions by first constructing a model for the electronically excited state,

including hyperfine structure. This is used to calculate the relevant energies and, together with the wavefunctions describing states F and G, the TDMs for the candidate transitions.

We make relevant codes available on GitHub ([https://github.com/arpitadas1/STIRAP\\_RbCs.git](https://github.com/arpitadas1/STIRAP_RbCs.git)), and it is also be available at the DOI of reference 62 of the revised manuscript.

Q2. In Section 2, could the authors briefly recall why only even partial waves are considered?

A2. The total parity is  $(-1)^L$  and is conserved in a collision, so only states with even values of  $L$  can cause resonances in s-wave scattering; scattering; values  $L = 0, 2, 4$ , etc. are indicated by labels s, d, g, etc. This text has been added to the revised manuscript in Section 2.

Q3. I found the description of Figure 1 in Section 2 quite pedagogical and relevant. However, it is sometimes a bit tedious to follow. Would there be a way to include more information in the Figure itself? For instance, would it be possible to number the consecutive avoided crossings, as it is quite tough to identify those which are very weakly avoided? Or may be to add arrows suggesting the path? In Panel a, the state d6 is not reported. Is it the same than in panel b? Would it be possible to label the unlabelled states, in particular in panel b, for completeness?

A2. We have strengthened the connection between the explanation in the main text and Fig. 1(a) by labelling the avoided crossings (i), (ii), (iii), and (iv). We believe that this will make the identification of which avoided crossings are being discussed more straightforward.

Our submitted manuscript incorrectly mentioned d6 (instead of d6') in the description of the panel (a) in Section 2, but this was a typo, for which we apologise.

The only state that appears in both panels is state s1. The steeply sloping states are all different and have been given distinct names without duplication. These names are abbreviated for ease of discussion, and because the states sd6 and ds6 are mixtures of the parent states s6 and d6. This is explained, and the full labelling of the states is described in the text of Section 2.

Q4. In Section 3, could the authors recall why the Feshbach resonance is mostly triplet Sigma u?

A4. The state F has mostly  $\sim a^3\Sigma^+$  character, because all the contributing states have relatively high spin projections,  $M_F = m_{f_{\text{Rb}}} + m_{f_{\text{Cs}}} \geq 3$ . We have added this text to Section 3 of the revised manuscript.

Q5. A bibliography issue: the A/b system has been also investigated in K\_2 [Eur. Phys. J. D 17, 319{328 (2001)], Li\_2 and Na\_2 [see references listed in the previous paper]

A5. Reference of K\_2 is added. References for Li\_2 and Na\_2, listed in the reference of the previous paper, are not for A-b, they are either A or b.

We have modified the text in Section 3 as "The system  $A^1\Sigma^+ - b^3\Pi$  has previously been investigated in many different alkali dimers [43–52]."

Q6. Commenting Table 1 the others wrote that "There is roughly a factor of two discrepancy between the calculated and measured values." As far as I can read, this concerns only the pump transitions. Am I correct? Furthermore, they wrote "our results may indicate that a more accurate electronic

wavefunction is required." If my previous remark is correct, how could we explain that the discrepancy occurs for the pump transition and not for the Stokes transition? Moreover, a factor of 2 on the TDM is huge, and usually, electronic wavefunctions are far more accurate to provide values better than a factor of 2. Or would the details of the spin-orbit coupling in the excited state (and thus the respective singlet and triplet fraction of the state) be the main source of uncertainty? I understand that the reason for this is not obvious, but this could be commented in more details, even if I understand that this does not change the conclusions of the paper. Could the experimental procedure to extract the TDM be also uncertain? I did not find in the paper how TDM values are extracted in the experiment, which is usually a quite delicate task.

A6. The experimental TDMs are obtained by measuring the Rabi frequency on each transition and normalising to the intensity of the light. The dominant source of uncertainty in the TDM is from the uncertainty in the intensity of the light. We believe that it is unlikely that these measured values could be incorrect by a factor of 2. Table 1 gives TDMs for the transitions at 181.5 G measured in Durham and Innsbruck, and the difference between these values gives a reasonable estimate of the uncertainties present in the experiments.

The differences between the experimental and theoretical values of the TDMs for the pump transitions are probably due to uncertainties in the electronic wavefunctions for the excited states. The calculated TDMs depend strongly on the electronic transition dipole functions, and this dependence is greater for the pump transitions because there is substantial oscillatory cancellation in the radial integrals.

We have added the above texts to our revised manuscript in Section 5 and some of the previous texts are modified by rewording "The values for the Stokes transitions are within about 50% of experiment, but there is roughly a factor of two difference between the calculated and measured values for the pump transitions".

### **Requested changes**

1. top of page 3: "We use unprimed, primed and double-primed quantum numbers for the Feshbach, excited and ground states [quantum numbers], respectively."

This sentence was correct as originally written: these are quantum numbers for states, and inserting an extra "quantum numbers" as we think is suggested would not make sense.

2. Figure 2, panel a: the asymptote of the excited state is labeled Rb(5S)+Cs(6P), but there are actually two asymptotes, presumably associated to 6P<sub>1/2</sub> and 6P<sub>3/2</sub>. I do understand that the space is limited, but it is an important information to specify that spin-orbit is included in this picture. In this respect the lower index of the Pi state should be clarified (i.e. Omega, defined in the main text)

Figure 2 has been modified accordingly in the revised manuscript.

## **Answers to the Reviewer 2**

We thank the reviewer for his/her review of our manuscript by recommending it for publication. He/she further posed a couple of valuable questions. We have tried to address all the queries.

Q1. In the introduction, it states, "Magnetoassociation following the new mixing protocol is straightforward on the much broader resonance at 352.7 G." Is this because the interspecies scattering length is zero at this field, or are there any other reasons? If there are other reasons, it might be helpful to elaborate more.

A1. The mixing is straightforward due to no interspecies scattering length at 355G. But at 352.7 G the scattering length is not zero.

For the magnetoassociation, the main reason is that we want to have a controlled magnetic field ramp, which is easier to perform if we don't have to make a large change to the field beforehand.

Q2. In section 7 on page 13, the manuscript mentions that the observed 100% Rabi contrast during microwave spectroscopy indicates a single hyperfine state occupancy. Could it still be possible that there is some occupancy of the  $(0,4)_0$  state, and this state also couples to a higher rotational state at a similar microwave frequency with a Rabi coupling similar to the  $(0,4)_1$  to  $(1,5)_2$  transition? I am wondering whether observing a 100% contrast is sufficient to claim single hyperfine state occupancy, although it is highly likely.

A2. This can indeed be an issue in some circumstances when probing rotational transitions in the molecules due to the many transitions available. However, we are confident in our ability to calculate the rotational and hyperfine structure of RbCs to 1 kHz precision in energy. We have found in our measurements that we can drive all of the population out of the ground state also on the transitions to  $(1,3)_2$  and  $(1,4)_3$ . For the microwave frequencies needed to drive these transitions there are no expected nearby transitions available from  $(0,4)_0$  as shown in Fig. 7(d,e). We believe that this is sufficient to claim single hyperfine state occupancy at the precision of current experiments.

To reflect this update, we have changed the text in Section 7 on Page 13 to read "...we find that we can drive Rabi oscillations on each of these strong transitions with 100% contrast; this indicates...".