Comment on "Microwave multiphoton transitions between Rydberg states of potassium"

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In a recent paper Stoneman, Thomson, and Gallagher [Phys. Rev. A **37**, 1527 (1988)] studied the excitation of potassium Rydberg atoms in microwave fields for a few values of the initial *S*-state principal quantum number. In one case the measured excitation probability as a function of microwave amplitude showed a rather broad feature that was explained in terms of level crossings between quasienergy states. The details of a crucial ingredient of this explanation, namely, a broadening mechanism, were, however, left unexplained.

Stoneman, Thomson, and Gallagher[1] report the observation of structure in the probability for exciting K ($n_0=19$) $^2S_{1/2}$ atoms to states of the adjacent $n=17$ $^2L>3$ manifold as a function of the amplitude of a 9.278-GHz microwave electric field. Such a structure was not observed by them for five other, nearby n_0 values. As a first step towards explaining the observed structure in terms of a multiphoton resonance, they also describe the results of a Floquet diagonalization on a limited basis consisting of the $n=17$ $L>1$, 19*S*, 19*P* atomic states together with 20 field Fourier components. However, because 20 photons are insufficient to bridge the gap between the 19*S* states and the $n=17$ manifold (which takes approximately 26 times the photon energy), such a calculation falls short of predicting the actual transition probability.

Instead, Stoneman, Thomson, and Gallagher[1] constructed the quasienergy level of the $|q_{19S,27}\rangle$ state by overlaying the graph of an ac Stark–shifted 19*S* state on the results of their Floquet diagonalization (their Fig. 14), and observe that the $|q_{19S,27}\rangle$ quasienergy crosses $|q_{n=17,0}\rangle$ Floquet manifold quasienergies at approximately 450 V/cm. ($|q_{19S,27}\rangle$ means quasienergy state $n=19$, $L=0$ with a relative photon index of 27.) The structure observed in their experiment and shown in their Fig. 4 is about 100 V/cm wide and is centered near 515 V/cm. Because Stoneman, Thomson, and Gallagher did not provide an absolute scale for the transition probability in this structure, we assume it is of order 1. Stoneman, Thomson, and Gallagher use their approach based on Floquet theory to give a general explanation (p. 1537 of Ref. 1) why the observed structure is inherently broad.

Motivated by our interest in microwave multiphoton transitions,[2] we have examined the approach based on Floquet theory used in Stoneman, Thomson, and Gallagher to explain their observations. In this Comment we report the results of our examination, concluding that their arguments do not suffice to explain why they observed a broad structure. Our work shows that the Floquet anticrossings contributing to their structure are well separated and would generally produce a sequence of very sharp peaks in their microwave excitation curve. To be observed as a single broad feature, it is necessary for them to be blended. Stoneman, Thomson, and Gallagher invoke transitions broadened by saturation (which we interpret as "power broadening") as an inherent mechanism for blending the peaks into a broad structure. Our work discussed here suggests that the blending is not inherent and that sharp, resolved peaks could be observed in K $n_0=19$ $^2S_{1/2}$ microwave excitation at 9.278 GHz. This Comment also explores possible explanations not considered by Stoneman, Thomson, and Gallagher[1] for why the structure observed by them was broad.

We start our analysis with the calculation of the Floquet states. Our atomic basis consisted of 20 states, namely $n=17$ $L=2,\ldots,16$; $n=18$ $L=1,2$; $n=19$ $L=0,1$; $n=20$ $L=0$.[3] The rationale for this selection was that this basis produced a faithful static field Stark map (as compared to a converged calculation on a larger atomic basis) and was at the same time still computationally managable. Full convergence of the Floquet diagonalization was reached with 131 field Fourier components. This number is more than *one order of magnitude* larger than the effective number (10) used by Stoneman, Thomson, and Gallagher, who included atom-photon states of either parity in their calculation. As was shown by Maquet, Shih-I Chu, and Reinhardt,[4] the Floquet basis needs only states of the same combined parity $(-1)^{L+k}$ as the initial state, where k is the field Fourier index (crudely, the photon number); other states are redundant. The computation of quasienergies for each field value took approximately 1 h of CPU time on a Digital Equipment Corporation VAX 8600 computer.

A section of the computed Floquet map is shown in Fig 1; it may be compared to Fig. 14 of Stoneman, Thomson, and Gallagher. Figure 1 shows a little more than one period of a spectrum computed for $M_L=0$. Because we are concerned with transitions out of an S state, the basis contains only states of even parity and the periodicity in Fig. 1 is $2\hbar\omega$ ($0.6189\ \mathrm{cm}^{-1}$). The spectrum shows very sharp and isolated anticrossings between the quasienergy state [indicated as (19S,30)] that join the initially excited state at zero field and ($n=17$ odd $L>3,3$) quasienergy manifold states. (A transition from the 19S state to an $n=17$ state in this region corresponds to a $30-3=27$ photon transition.) In agreement with Stoneman, Thomson, and Gallagher, the computed anticrossings cluster in a region near 450 V/cm. This region is marked in Fig. 1. Figure 14 of Stoneman, Thomson, and Gallagher shows results of a Floquet diagonalization carried out for only seven field values equally spaced between 0 and 600 V/cm. This is too coarse a grid to reveal details described here.

In the relevant cluster of anticrossings around 450 V/cm we chose the grid of field points fine enough to permit determination of anticrossing widths. The smallest grid spacing used is 0.1 V/cm. Outside this region, where the grid is much coarser, Fig. 1 shows many apparent anticrossings that are not real. The computed Rabi periods at the three largest (rightmost in the region indicated in Fig. 1) anticrossings ranged from 2.5×10^{-9} to 2.5×10^{-8} s. These anticrossings give rise to very narrow peaks in the time-averaged transition probability; all respective widths are smaller than 1 V/cm, compared to their typical spacing of 10 V/cm. Stoneman, Thomson, and Gallagher did not observe sharp peaks in their experiment.

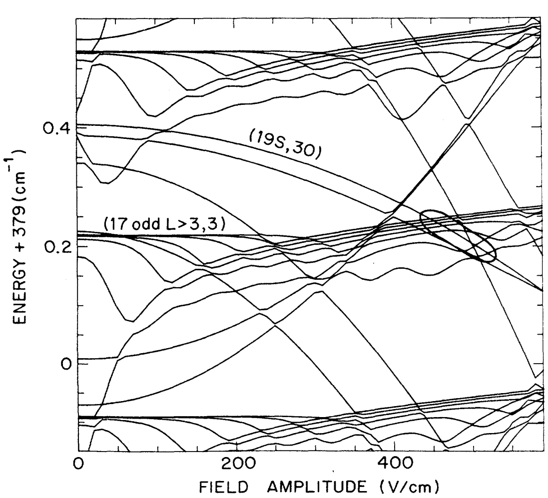

FIG. 1. Map of quasienergy levels of excited potassium $n=19$ $^2S_{1/2}$ atoms in a 9.278-GHz microwave electric field. The adiabatic initial state is indicated as (19S,30). Anticrossings mediating $n_0=19$ $^2S_{1/2}\rightarrow n=17$ $^2L_{1/2}$, $L>3$ transitions are encircled (see text).

In our calculation we have ignored fine-structure interaction, as have Stoneman, Thomson, and Gallagher. This is not justified; for example, the energy difference at zero field between the $17\,^2P_{1/2}$ and the $17\,^2P_{3/2}$ levels is about half the microwave photon energy. Although their shifts at zero field amplitude are much smaller, the high-angular-momentum states at large amplitudes will be mixed with states of lower angular momentum, and thus will also acquire shifts due to spin-orbit interaction. We expect that inclusion of the $J=\frac{3}{2}$ states in the calculation will double the number of peaks in the transition probability as a function of field amplitude, but the full set of peaks should still be separated by significantly more than their width.

Therefore, we now seek mechanisms that would cause the influences of the separated Floquet anticrossings to be blended together. A requirement is that such a mechanism should broaden each resonance, while not decreasing its height. Power broadening is naturally included in the Floquet calculation. Therefore, if we have correctly interpreted "transitions broaden[ing] to saturation" (Ref. 1, p. 1537) as power broadening, we would like to point out that Stoneman, Thomson, and Gallagher should not use this to explain the blending of transitions due to separated Floquet anticrossings.

In the microwave multiphoton excitation experiments of Stoneman, Thomson, and Gallagher, the transitions were driven with temporal pulses of microwaves.[5] When a pulse is produced only by switching on and off a traveling-wave-tube pulse amplifier driven by a cw source, it is a well-known fact in microwave engineering that the properties of the pulse-amplified output are strongly affected by transients near turn-on and turn-off.[6] These transients produce what is commonly referred to as "phase chirp." We are aware of no discussion of its influence on the high-order multiphoton transitions considered by Stoneman, Thomson, and Gallagher and in this Comment. However, in two-photon laser spectroscopy this problem is currently being investigated both theoretically and experimentally.[7] These studies have shown that apparent line centers and line shapes of two-photon transitions can be strongly affected by pulse-amplifier transients, such as phase chirp. What is perhaps surprising is that this can be so even if the pulse-amplified radiation is filtered by a cavity. For higher-order multiphoton transitions these effects will also likely be important and will have an even more complicated dependence on the parameters describing the details of the pulse.

We would expect that the effects of the pulse-amplified transients would be very much dependent on how the pulsed microwave experiment is operated, a situation open to experimental test.

Phase chirp is but one way to parametrize the lack of perfect coherence in a field. Coherence, specifically a coherence time, figures importantly in how Stoneman, Thomson, and Gallagher make an incoherent sum of transition probabilities over the pulse duration. Specifically, they stated a coherence time of 1 ns and, with their interaction time being 1 μs, implied that small transition probabilities should be multiplied by a factor of 10^3. Such a summation gives rise to *homogeneous* broadening and leads us to try to understand the source of the 1-ns coherence time stated by Stoneman, Thomson, and Gallagher. In this respect they have mentioned inhomogeneities of the microwave electric field; the static field is nominally zero in the experiment on which we focus here. In their atomic beam, the average atom moved less than 1 μm during their stated coherence time. The atoms were about 1 mm away from the nearest cavity wall, which is about 10^3 times their traveling distance. Therefore, the incoherence induced by the spatial field inhomogeneity that is experienced by a *single* atom in 1 ns should be negligible. We believe, instead, that a signal consisting of the integrated contributions of many individual atoms provides a more enlightening picture: each atom interacts coherently for a time longer than 1 ns but a different field strengths (inhomogeneous broadening).

The distinction between homogeneous and inhomogeneous broadening and a discussion of their causes is the key point of our argument. Let it be clear that the spatial inhomogeneity of the microwave field may give rise to both types of broadening. Roughly, to quantify inhomogeneous broadening one needs the distribution of fields seen by different atoms traversing the interaction region, whereas for homogeneous broadening it is also necessary to specify how fast the field amplitude fluctuates on each particle's path.

Inhomogeneous broadening is expressed as a convolution of the resonance profile with the field-amplitude distribution. The broadening of each resonance is then accompanied by a reduction of its height. The spatial microwave field inhomogeneity in the experiment of Stoneman, Thomson, and Gallagher is at least the variation of the cavity mode over the interaction region. At a nominal field amplitude of 500 V/cm this variation would be 10 V/cm. Resonances with an intrinsic width of less than 1 V/cm would therefore be reduced in strength to less than 0.1. However, Stoneman, Thomson, and Gallagher observe a single broad resonance with strength of order unity (p. 1530 of Ref. 1). Therefore, spatial inhomogeneous broadening cannot explain the occurrence of a single broad structure observed by Stoneman, Thomson, and Gallagher.

A microwave field that fluctuates in time is another source of broadening. Fluctuations can occur in amplitude, phase, and frequency. Above we referred to the case of phase chirp. We will now make some admittedly simple semiquantitative estimates of the homogeneous broadening that would be caused by amplitude fluctuations. A natural scale of these fluctuations is the extent ΔF of an individual anticrossing, here of order 1 V/cm, relative to the anticrossing field $F \simeq 500$ V/cm.[8] Thus, $\Delta F/F \simeq 2\times10^{-3}$. The most natural time scale for a cavity resonator is its field "ringdown" time $\tau_r = 2Q/\omega$ ($=4\times10^{-8}$ s, as implied by the quality factor $Q=1100$ quoted by Stoneman, Thomson, and Gallagher). Field fluctuations inside the cavity due to source or amplifier noise are strongly damped on this time scale. Of course, the actual magnitude of these fluctuations on any given time scale depends on how hard they are being driven. The Lorentzian shape of the cavity response function dictates that input noise fluctuations in the microwave power with a characteristic (coherence) time τ_c are reduced by a factor $1/(1+\tau_r^2/\pi\tau_c^2)$.

Assume that the field fluctuations cause Rabi transitions in τ_c with a probability averaged over τ_c,

$$p = \left\{ 2 \left[1 + \left[\frac{F'}{U} \frac{d\Delta E}{dF} \right]^2 \right] \right\}^{-1} , \tag{1}$$

where F' is the difference between the averaged field amplitude and the anticrossing field, U the level separation at $F'=0$, and $d\Delta E/dF$ the differential slope of the quasienergy levels. The use of the averaged probability requires that the Rabi period be no larger than τ_c ($=4\times10^{-8}$ s, see above). Next assume that the phase of a given atomic wave function is completely randomized between subsequent transitions. The final transition probability after an interaction time τ_i ($=1\times10^{-6}$ s) would then be given by

$$P = \tfrac{1}{2}(1 - e^{-2p\tau_i/\tau_c}) . \tag{2}$$

With the numbers quoted above, this simple model requires a noise level of about 1% of the microwave power coupled into the cavity to produce a broadening of the peaks to about 5 V/cm. These arguments can, of course, at most establish an order of magnitude for broadening effects due to noise. We point out that a theory of transitions in a noisy radiation field must involve the precise statistical properties of this noise.[9]

Finally, as we have discussed elsewhere,[2] we expect that dynamic effects due to the microwave pulse shape significantly affect the shape and width of these multiphoton resonances. To model this requires detailed knowledge about the shape of the microwave pulse. Stoneman, Thomson, and Gallagher did not describe the shape of their pulse.

In conclusion, we were led to this Comment because a satisfactory broadening mechanism was not presented by Stoneman, Thomson, and Gallagher, a mechanism that could lead from the situation of sharp structures in our own experiments with He atoms[2] to a broad structure in the experiments by Stoneman, Thomson, and Gallagher on K atoms.[1] Our estimates lead to the conclusion that the broadening will only be understood if one has detailed information about the coherence properties of the microwave field actually seen by the atoms. In experiments investigating multiphoton transitions driven by pulsed fields, therefore, it is necessary that as much detailed information as possible be given about the properties of the pulsed fields.

[1]R. C. Stoneman, D. S. Thomson, and T. F. Gallagher, Phys. Rev. A **37**, 1527 (1988). To avoid possible confusion, notice that these authors report two different kinds of experiments: (i) one that used a static field superimposed with the microwave field to tune the energy-level separations (see, e.g., the data of their Fig. 5) and (ii) a setup that used only a microwave field, though, of course, stray static fields are always present. This Comment focuses on the physics of (ii).

[2]W. van de Water, K. A. H. van Leeuwen, S. Yoakum, E. J. Galvez, L. Moorman, B. E. Sauer, and P. M. Koch, Phys. Rev. Lett. **63**, 762 (1989); W. van de Water *et al.*, Phys. Rev. A **42**, 572 (1990).

[3]We used the following values for the quantum defects: $\mu_{L=0}=2.180$, $\mu_1=1.712$, $\mu_2=0.277$, $\mu_3=0.010$, whereas a polarization formula with core polarizability 5.49 was used for the quantum defects with $L>3$. See C. -J. Lorentzen and K. Niemax, Phys. Scr. **27**, 300 (1983). This set of quantum defects differs from that used by Stoneman, Thomson, and Gallagher (Ref. 1), who quoted numbers with one decimal place less and ignored quantum defects of states with $L>2$. The use of these two different sets results in quasienergy maps that differ significantly. For example, with the set of quantum defects given above, we found that the computed anticrossings occur at field amplitudes that are 30 V/cm higher than those we computed from the set of quantum defects used by Stoneman, Thomson, and Gallagher, whereas the computed anticrossing widths are typically a factor of 2 larger.

[4]A. Maquet, Shih-I Chu, and W. P. Reinhardt, Phys. Rev. A **27**, 2946 (1983).

[5]There is an important difference in how the microwave pulses were produced in Ref. 1 as compared to the He experiments in Ref. 2. In the K experiments of Ref. 1, the microwave pulses were produced by actually switching in time. In the He experiments the microwaves were fed continuously in time into the resonant cavity. From the perspective of the He atoms going through the cavity at constant velocity, the spatial variation of the microwave amplitude translated to a temporal variation in their rest frames. Therefore, the properties of the microwave field are in this case determined by the characteristics of a continuous source and amplifier driving a resonant cavity.

[6]John van Dyke (private communication). A standard way to greatly reduce these transients is to run in a quasi-cw mode with use of another low-power switch. Only after the pulse amplifier is switched on and has settled does one open the switch between it and the source. Similarly one closes the switch between it and the source before the pulse amplifier is turned off. This quasi-cw mode is called "pulse pedestalling" in microwave engineering.

[7]K. Danzmann, M. S. Fee, and S. Chu, *Laser Spectroscopy IX*, edited by M. S. Feld, J. E. Thomas, and A. Mooradian (Academic, San Diego, 1989), pp. 328–331; K. Danzmann (private communication).

[8]In another context, R. Blümel, R. Graham, L. Sirko, U. Smilansky, H. Walther, and K. Yamada, Phys. Rev. Lett. **62**, 341 (1989) showed that in an experiment with Rb Rydberg atoms driven by a sinusoidal field and broadband noise, the time required for noise-induced scrambling of quantal phase difference was inversely proportional to the noise power.

[9]A recent study of radiation noise in the framework of the rotating-wave approximation was reported by R. Boscaino and R. N. Mantegna, Phys. Rev. A **40**, 5 (1989); **40**, 13 (1989); and **40**, 2217 (1989); Opt. Commun. **73**, 289 (1989); J. Opt. Soc. B (to be published). See also references therein.