

Fine-structure intervals of f states in highly excited Na^{\dagger}

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(Received 22 March 1977)

The fine-structure intervals of the $\text{Na } 11f, 13f,$ and $14f$ states have been measured by a microwave-resonance method yielding values of 10.52(21), 6.34(12), and 5.02(8) MHz, respectively. These values are $\sim 5\%$ lower than the theoretical values for the corresponding hydrogenic states. We have also investigated the use of Ramsey's method of separated oscillatory fields as a means of obtaining narrower resonances.

INTRODUCTION

It is well known that the fine-structure (fs) intervals of $\text{Na } p$ and d states are very different from the hydrogenic p and d fine-structure intervals.^{1,2} However, the measurement of the $\text{Na } 4f$ fs interval by Liao and Bjorkholm³ and our previous measurements⁴ of the $\text{Na } 11f$ - $17f$ fs intervals have not, within the experimental uncertainties, shown any departure from the hydrogenic values. The $\text{Na } p$ and d fs intervals are so different from H, due to the perturbing effects of the Na^+ core, that it seems likely that in the $\text{Na } f$ states perturbing effects will persist with the result that the $\text{Na } f$ -state fs will not be exactly hydrogenic. The departures of the $\text{Na } f$ -state fs interval from hydrogenic theory, which we shall term the Na-H difference, do not appear to originate from the electrostatic polarization or penetration of the Na^+ core by the valence f electron. Thus, the Na-H difference must arise from quantum-mechanical effects of the electrons of the Na^+ core and their interaction with the valence electron, and as a consequence, the Na-H difference is a sensitive probe of these effects. Accordingly, we set out to improve the accuracy of our previous measurements of the $\text{Na } 11f, 13f,$ and $14f$ fs intervals.

EXPERIMENTAL METHOD

These experiments are, in principle, the same as those described in Ref. 4. The basic idea of the experiment is illustrated by Fig. 1, which shows the relevant energy levels of the $n=13$ level. Two pulsed dye lasers of 4ns pulse duration are used to pump the atoms in a stepwise fashion from $3s_{1/2}$ to $3p_{3/2}$ then to $13d_{5/2}$. A microwave field is used to drive the $d_{5/2}$ - $f_{7/2}$ and $d_{5/2}$ - $f_{5/2}$ transitions, and we detect the transitions by observing the infrared (ir) $13f$ - $3d$ and $3d$ - $3p$ fluorescence. When the microwave frequency is not at the resonance frequency of either of the two transitions indicated in the inset of Fig. 1,

there is no infrared fluorescence, but as the frequency is swept through the resonances sharp increases in the fluorescence are observed, as shown in Fig. 2.

Since the apparatus is described in detail elsewhere,⁴ we only describe its basic features and explain the modifications made for these experiments. The Na vapor is contained in a cylindrical Pyrex cell, which we kept at a temperature of 160°C , producing a Na pressure of 1.5×10^{-5} Torr. The dye-laser beams pass along the axis of the cell, and the microwaves are introduced from a direction perpendicular to the axis by placing a horn as close to the side of the cell as possible. The ir fluorescence was detected by a photomultiplier placed on the opposite side of the cell from the microwave horn.

Our previous measurements were done unshielded from Earth's magnetic field, which led to linewidths of ~ 3 MHz. For these experiments we have put a double magnetic shield around the cell. The inner shield is made of 0.010-in.-thick mu-metal, and the outer shield is made of 0.030-in.-thick mild steel. As we shall see, these magnetic shields reduced the Zeeman broadening to a negligible level.

In the absence of Zeeman or Stark broadening, the minimum linewidth of an observed resonance is determined by the uncertainty principle to be the inverse of the time the atoms are exposed to the microwave field. Thus, to obtain the narrowest possible linewidth we should delay the observation of the ir fluorescence from the f state as long as possible after the laser pulses. However, because of the radiative decay of the d and f states, the signal strength declines exponentially with the delay in the observation time.⁵ Thus we must compromise between linewidth and signal strength. Since the lifetimes of these d and f states are 1-2 μs , we have delayed the observation of the ir fluorescence for 2 μs after the laser pulses. Specifically the observation time is a 0.5- μs interval centered 2 μs after the laser pulses. This observation scheme has the advantage of producing

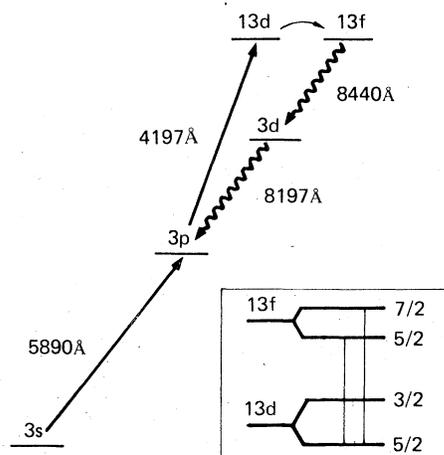


FIG. 1. Energy levels for the experiment on the Na $n=13$ state. The upward straight arrows indicate the two laser pumping steps. The downward wavy arrows indicate the observed fluorescent decays and the curved arrow indicates the microwave $d-f$ transition. The inset shows the $13d$ and $13f$ levels, and the two vertical lines show the two transitions emanating from the $d_{5/2}$ state.

the maximum resonance signal when the microwave power is set to induce the $d \rightarrow f$ transition in $2 \mu\text{s}$. Thus we have both the maximum signal and the uncertainty-principle linewidth of 0.5 MHz at the same setting of the microwave power.

Since the point of these measurements is to determine the Na-H difference, small frequency shifts of $\sim 0.1 \text{ MHz}$ are important and some care must be taken to ensure that there is not a systematic shift in our observed frequencies. The three most likely sources of shifts are pressure shifts, Stark shifts, and Zeeman shifts. The background gas pressure was $\sim 10^{-6} \text{ Torr}$; so the most likely source of pressure shifts is Na. To look for pressure shifts, we raised the temperature of the cell to 185°C , corresponding to a Na pressure of $7.5 \times 10^{-5} \text{ Torr}$. At this point we observed noticeable ($\sim 0.5 \text{ MHz}$) broadening of the lines but no apparent shift; consequently, we have concluded that under the usual operating conditions pressure effects were unimportant. There is no direct way of checking the Stark or Zeeman shifts, but the linewidth of the observed resonances serves as an indirect check. As shown by Fig. 2, the linewidth (FWHM) of the resonances is 0.5 MHz , the inverse of the transition time of $\sim 2 \text{ MHz}$; that is, we obtained a linewidth corresponding to the uncertainty-principle limit. This suggests that there is no observable ($<0.1 \text{ MHz}$) broadening of the transition due to other effects such as electric or magnetic fields. Since we drive transitions of all the m_j levels, any electric or magne-

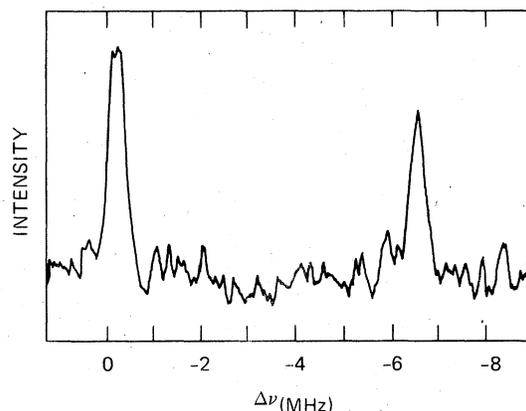


FIG. 2. $13d_{5/2}-13d_{7/2}$ and $13d_{5/2}-13d_{5/2}$ resonances on a sweep of decreasing frequency. The microwave power was increased by 15 dB , 2 MHz before the $13d_{5/2}-13d_{5/2}$ resonance was reached.

tic field would be manifested as a broadening. For both magnetic and electric fields, we estimate that the shifts in the frequencies are less than 20% of the broadenings so that the shifts are $<20 \text{ kHz}$, which is not a limiting factor in these experiments.

The last important change was in the method used to measure the microwave frequency. To drive the $d-f$ transitions, we used a swept-frequency oscillator which we beat against a fixed-frequency local oscillator to produce a beat note at $\sim 30 \text{ MHz}$, which we counted. Thus, when we swept the first oscillator through the two resonances, the beat-frequency counter told us their frequency separation. In this way we were able to measure the difference between the $d_{5/2}-f_{5/2}$ and $d_{5/2}-f_{7/2}$ transition frequencies (the f fs interval) with great accuracy, although we did not know the absolute frequencies. The major limitation to this technique is the drift of the local oscillator during the $2-3 \text{ min}$ required for the frequency sweep. Over this time period the drift was measured to be $<25 \text{ kHz}$, which was not a limitation in these experiments.

The electric dipole matrix elements for the $\Delta J = 1$ transitions are roughly a factor of 5 larger than the $\Delta J = 0$ transitions. Thus, we found it necessary to alter the microwave power by 15 dB in the middle of each sweep to avoid power broadening the $d_{5/2}-f_{7/2}$ transition. For example, in the $n=13$ sweep shown in Fig. 2, we estimate that the incident powers for the $d_{5/2}-f_{7/2}$ and $d_{5/2}-f_{5/2}$ resonances were 50 nW/cm^2 and $1.5 \mu\text{W/cm}^2$, respectively.

Among the other improvements aimed at narrowing the resonances, we applied Ramsey's method of separated oscillatory fields.⁶ Ramsey's

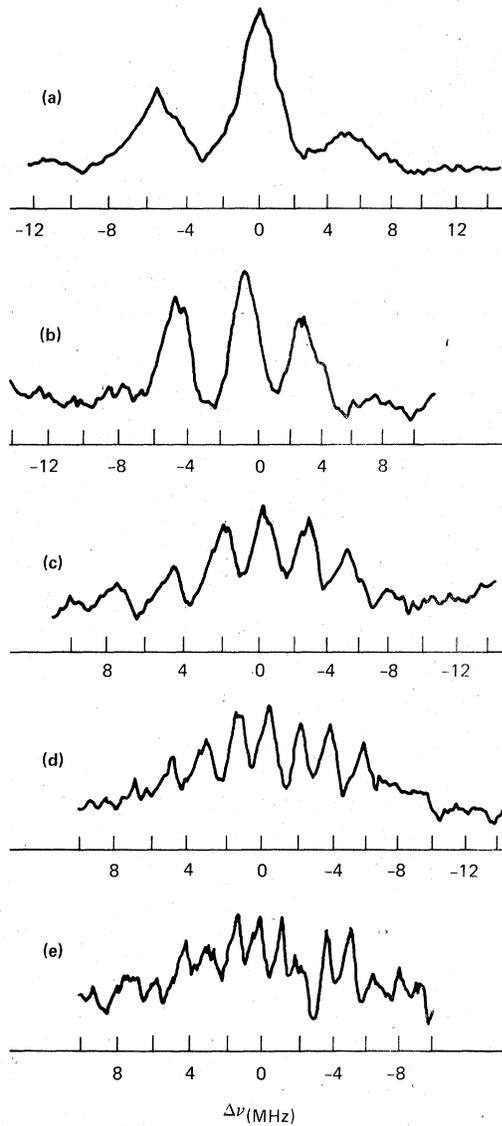


FIG. 3. Ramsey interference patterns of the $13d_{3/2}$ - $13f_{5/2}$ transition at 39.203 GHz. Two pulses of microwave power $0.1 \mu\text{s}$ long were applied which were separated by (a) $0.1 \mu\text{s}$, (b) $0.15 \mu\text{s}$, (c) $0.4 \mu\text{s}$, (d) $0.55 \mu\text{s}$, and (e) $0.75 \mu\text{s}$.

method of spatially separated oscillatory fields is commonly used in high-precision radio-frequency molecular-beam experiments. In its most familiar application there are two spatially separated oscillatory fields. However, the use of spatially separated fields is impractical for rapidly decaying excited states, such as we have in these experiments. Consequently, we have used temporally rather than spatially separated oscillatory fields. Temporally separated fields have been used less frequently than spatially separated fields, perhaps the most elegant application being the recent optical work of Hänch *et al.*⁷ To use Ramsey's method, we used two microwave pulses $\sim 0.1 \mu\text{s}$ long separated by a time T which we varied from $0.1 \mu\text{s}$ to $0.75 \mu\text{s}$. The first pulse began within 50 ns after the laser pulses, and we detected the fluorescence for $0.5 \mu\text{s}$ after the end of the second microwave pulse. In the $n=13$ experiments shown in Fig. 3 microwave power at 39.2 GHz was produced by doubling the output of a 19.6-GHz oscillator with a diode. Normally the diode was reverse biased and no 39.2-GHz power was generated. When the diode was forward biased with a pulse generator, we obtained pulses of 39.2-GHz power. The microwave power had a rise time of 20 ns, constant power for 75 ns, and a fall time of 20 ns, exactly following the waveform of the pulse generator. In Fig. 3 we show the Ramsey interference patterns for the $13d_{3/2}$ - $13d_{5/2}$ transitions at 39.2 GHz. There are asymmetries in the pattern which we attribute to 10–20% fluctuations in the microwave power over the 20-MHz range swept. Although Ramsey's method gave us narrower resonances, we did not use it in these f fs measurements for two reasons. As is apparent from Fig. 3, it was not always obvious which peak was the central peak, and the resonance for the $d_{5/2}$ - $f_{5/2}$ and $d_{3/2}$ - $f_{5/2}$ transitions would have overlapped badly.

RESULTS AND DISCUSSION

The results of the Na f fs measurements are given in Table I with the hydrogenic values.⁸ The

TABLE I. Na f -state fine-structure intervals.

n	$d_{5/2}$ - $f_{7/2}$ frequency ^a (MHz)	Na f fs interval (MHz)	H fs interval ^b (MHz)	% difference from H
11	64 399.6(8)	10.52(21)	10.98	-4.2(21)
13	39 253.4(12)	6.34(12)	6.65	-4.7(19)
14	31 493.9(6)	5.02(8)	5.35	-5.2(16)

^aSee Ref. 4.

^bSee Ref. 8.

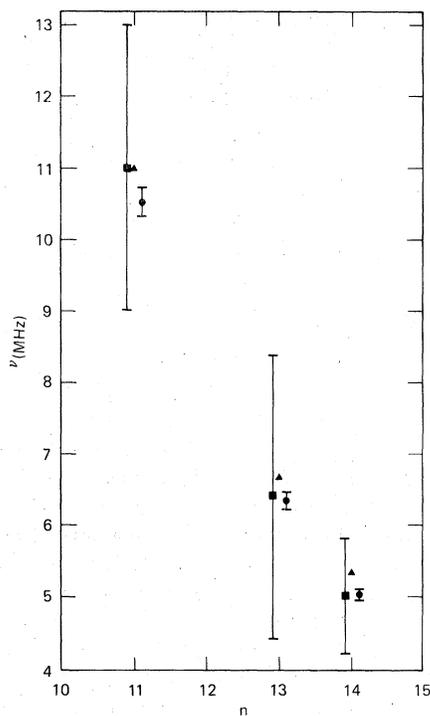


FIG. 4. Plot of the hydrogenic f fs interval (▲), the measurements of Ref. 4 (■), and the results of this work (●).

uncertainties reflect the statistical uncertainties and our estimates of systematic effects. For reference the absolute $d_{5/2}-f_{7/2}$ frequencies are also given. In Fig. 4 these and the previous Na f fs measurements are plotted with the hydrogenic values. As shown by Fig. 4, these microwave measurements of the Na f fs intervals lie 4–5% below the hydrogenic values, in contrast to Liao

and Bjorkholm's optical measurement of the Na $4f$ fs interval, 229(4) MHz, which is in agreement with the hydrogenic value of 228 MHz. At the moment the two measurements appear to disagree. Experiments are currently in progress to measure the Na $4f$ fs interval by resonance techniques which should clarify the situation.⁹

As mentioned previously, the Na-H difference cannot be explained by core polarization alone. The valence electron polarizes the core, which in turn produces an increase in the electric field at the valence electron, increasing the fine-structure interval. However, distortion of the core produces a net circulating current which produces a magnetic field at the valence electron which exactly cancels the electrostatic increase in the fs interval due to the dipole polarizability.¹⁰ Quadrupole and higher moment polarizabilities would cause very small increases in the fs interval, as would core penetration effects. Thus, the observed negative Na-H difference must arise predominantly from exchange core polarization effects.¹¹ Comparison of our result of -5% for the Na-H difference with preliminary theoretical calculations based on exchange core polarization effects¹² shows qualitative agreement. We hope that these measurements will stimulate renewed experimental and theoretical interest in the problem of the fine structure of the alkali atoms.

ACKNOWLEDGMENTS

We would like to acknowledge helpful discussions with R. R. Freeman, M. L. Zimmerman, G. A. Victor, and R. M. Sternheimer. We gratefully acknowledge the loans of microwave equipment by S. Tetenbaum and J. Watgen and of a counter by W. Bichel.

[†]Work by the U.S. Air Force Office of Scientific Research under Contract No. F44620-74-C-0069.

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