Differential Stark effect in the ground-state hyperfine structure of gallium

Stanley J. Rosenthal
Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
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An atomic-beam magnetic-resonance technique has been used to measure the differential Stark splitting between "flop-in" Zeeman levels in the ground-state hyperfine structure of naturally occurring gallium. Precision measurements of the shift were made at several electric fields up to 200 kV/cm and various rf power levels. A significant rf dependence was observed, and it was necessary to extrapolate to zero rf power to obtain the final result. The shift was determined to be $500(16)\times10^{-10}$ Hz/(V/cm)^2, including systematic uncertainties in the electric field value and the filling factor. This can be interpreted as a tensor polarizability of $667(21)\times10^{-10}$ Hz/(V/cm)^2 or $2.700(0.84)\times10^{-4}a_0^3$. The shift increases the magnetic splitting between the $M_F=-1$ and the $M_F=-2$ sublevels. The theoretical and experimental results are compared and found to agree well to within the stated errors.

I. INTRODUCTION

The change in the ground-state hyperfine-structure splitting in a free atom produced by a uniform electric field was first observed in cesium by Haun and Zacharias.[1] The smaller shift induced by an electric field between Zeeman levels within a given ground state-hyperfine level was also first observed and measured in cesium by Lipworth and Sandars[2] and measurements extended to the other alkalis by Gould *et al.*[3] and Carrico *et al.*[4] Precision measurements of the shift in the alkali metals were later performed by Mowat.[5]

A detailed theory of the effect in $^2S_{1/2}$ ground-state atoms has been given by Sandars[6] and later extended by Angel and Sandars[7] and Khadjavi and Lurio[8] to atoms with $^2P_{1/2}$ ground states. Angel, Sandars, and Woodgate[9] have measured the shift in $^2P_{1/2}$ ground-state aluminum and found agreement with theory.

Here we report results on measurements in the $^2P_{1/2}$ state of naturally occurring gallium. Theoretical values for the polarizability of the two naturally occurring isotopes are computed, and an analysis of the net effect of the two Ramsey patterns on the apparent resonance is presented.

II. THEORETICAL CONSIDERATIONS

It is useful to define the tensor polarizability[7,8] α_t by

$$-\tfrac{1}{4}\alpha_t(J,F)=\langle J,F,F\,|\mathcal{H}^{\rm ten}_{\rm eff}|J,F,F\rangle/(3E_z^2-E^2)\,, \tag{1}$$

where $|J,F,F\rangle$ is the stretched Zeeman state and $\mathcal{H}^{\rm ten}_{\rm eff}$ is the tensor part of the effective Stark Hamiltonian as derived in Refs. 7 and 8. Then the Stark shift of any substate is related to the uniform electric field applied along the axis of quantization z by

$$\Delta W(F,M)=-\tfrac{1}{4}\alpha_t(J,F)\frac{3M^2-F(F+1)}{F(2F-1)}(2E^2)\,. \tag{2}$$

In the special case of $J=\frac{1}{2}$ the theoretical value of α_t vanishes identically in the absence of hyperfine structure terms which can mix in states of higher J value.[7] The calculation including these terms leads to small but finite tensor polarizability. Sandars[9] shows that for a $J=\frac{1}{2}$ level the combination in second-order perturbation theory of the tensor Stark operator and the hyperfine structure operator between doublet states leads to an energy shift $W(F,M)$ of the form

$$\Delta W(F,M)=2\frac{\langle {}^2P_{1/2},F,M\,|\mathcal{H}^{\rm hfs}|\,{}^2P_{3/2},F,M\rangle\langle {}^2P_{3/2},F,M\,|\mathcal{H}^{\rm ten}_{\rm eff}|\,{}^2P_{1/2},F,M\rangle}{W_{1/2}-W_{3/2}}\,. \tag{3}$$

From Eq. (1), we have

$$\Delta W(F,F)=-\tfrac{1}{2}\alpha_t(J=\tfrac{1}{2},F)E^2\,. \tag{4}$$

In the LS-coupling approximation (which can be shown to introduce no significant errors), both $\alpha_t(J=\frac{3}{2})$ and $\langle {}^2P_{3/2}|\mathcal{H}^{\rm ten}_{\rm eff}|\,{}^2P_{1/2}\rangle$ are proportional to the reduced matrix element $\langle L\|\mathcal{H}^{\rm ten}\|L\rangle$ and related directly to each other. For the values $F=2$, $I=\frac{3}{2}$, $J=\frac{1}{2}$, $J'=\frac{3}{2}$, we find

$$\langle SLIJ'FF\,|\mathcal{H}^{\rm ten}|\,SLIJFF\rangle=-(2/\sqrt{7})[\tfrac{1}{2}\alpha_t(J'=\tfrac{3}{2})E^2]\,. \tag{5}$$

The hyperfine interaction term in Eq. (3) is evaluated similarly by expressing all elements of the hyperfine matrix in terms of the same angular momenta and spherical tensor operators.

The diagonal terms are proportional to the hyperfine-structure constants. The value of these constants, $A_1(J)$ and $A_2(J)$, are derived from measurements in Table I. Thus we can express the off-diagonal terms as a linear combination of these.

First we express $\mathcal{H}^{\rm hfs}$ as the sum of products of tensors in the electronic and nuclear spaces[12]

$$\mathcal{H}^{\rm hfs}=\sum_k \underset{\sim}{\vec{T}}{}^k_e\cdot\underset{\sim}{\vec{T}}{}^k_n, \tag{6}$$

where only $k=1,2$ are significant.

We make use of the expression from Sandars and Beck[13] for the magnetic-dipole operator

$$\underset{\sim}{\vec{T}}{}^1_e=2M_0[\langle r_l^{-3}\rangle\underset{\sim}{\vec{L}}-\sqrt{10}\langle r_{sc}^{-3}\rangle\{\underset{\sim}{\vec{S}}\cdot\underset{\sim}{\vec{C}}{}^2\}^1+\langle r_s^{-3}\rangle\underset{\sim}{\vec{S}}]. \tag{7}$$

The three relativistic radial integrals are not evaluated explicitly for gallium, but from Schwartz[12,14] we obtain the useful ratio

$$\langle r_l^{-3}\rangle/\langle r_{sc}^{-3}\rangle=0.916. \tag{8}$$

Also

$$A_k(J)=\langle I,J,F=I+J,M=F\,|\,\underset{\sim}{T}{}^k_e\cdot\underset{\sim}{T}{}^k_n\,|\times I,J,F=I+J,M=F\rangle. \tag{9}$$

Hence two more independent equations are obtained in terms of the constants $A_1(\frac{1}{2})$ and $A_1(\frac{3}{2})$. The quadrupole operator is $\underset{\sim}{T}{}^2_e=-e\langle r_Q^{-3}\rangle C^2$, which may be evaluated from $A_2(\frac{3}{2})$. We now insert in Eq. (3) the following for gallium:

$$\langle\tfrac{3}{2},\tfrac{1}{2},2,2\,|\,\mathcal{H}^{\rm hfs}\,|\,\tfrac{3}{2},\tfrac{3}{2},2,2\rangle=-0.812A_1(\tfrac{1}{2})+1.062A_1(\tfrac{3}{2})-2A_2(\tfrac{3}{2}). \tag{10}$$

The known fine-structure separation[15] is

$$W_{1/2}-W_{3/2}=-826.24\ \text{cm}^{-1}=-2.479\times10^7\ \text{MHz}. \tag{11}$$

We make use of the value of $\alpha_t(J=\frac{3}{2})$ measured by Petersen[16] to be

$$\alpha_t(J=\tfrac{3}{2})=-2.76(4)\times10^{-3}\ \text{Hz/(V/cm)}^2. \tag{12}$$

The resulting theoretical values of $\alpha_t(J=\frac{1}{2})$ from Eqs. (3) and (5) are for ^{69}Ga,

$$\alpha_t(\tfrac{1}{2})=658(10)\times10^{-10}\ \text{Hz/(V/cm)}^2=2.66(4)\times10^{-4}a_0^3, \tag{13}$$

and for ^{71}Ga,

$$\alpha_t(\tfrac{1}{2})=802(12)\times10^{-10}\ \text{Hz/(V/cm)}^2=3.25(5)\times10^{-4}a_0^3. \tag{14}$$

TABLE I. Hyperfine interaction constants and off-diagonal matrix elements.

Constant		^{69}Ga (MHz)	^{71}Ga (MHz)
A[a]	$(J=\frac{1}{2})$	1338.99	1701.35
$A_1(\frac{1}{2})$		1004.25	1276.01
A[b]	$(J=\frac{3}{2})$	190.794	242.434
B[b]	$(J=\frac{3}{2})$	62.522	39.399
$A_1(\frac{3}{2})$		429.28	545.47
$A_2(\frac{3}{2})$		15.63	9.85
$\langle\frac{3}{2},\frac{1}{2},2,2\,\vert\,\mathcal{H}^{\rm hfs}\,\vert\,\frac{3}{2},\frac{3}{2},2,2\rangle$		−390.80	−476.53

[a] Reference 10.
[b] Reference 11.

III. METHOD

An atomic-beam magnetic-resonance apparatus was aligned to observe the flop-in transition, $(F=2,M=-1)\leftrightarrow(F=2,M=-2)$, in gallium. The interaction region contained a pair of heated glass electric field plates, between the loops of a Ramsey double-hairpin structure. The loops and plates were in a uniform weak magnetic field produced by Helmholtz coils. The electric and magnetic fields were parallel to 0.02 deg.

The frequency of the flop-in transitions were observed in magnetic fields of 740 and 375 mG at various electric field strengths from 60 to 200 kV/cm as a function of the rf power applied to the loops.

The linewidth of the transition was 380 Hz, and the shifts ranged from 140 to 1600 Hz. The overall linewidth of the resonance envelope was 68 KHz. Slope detection[3] provided a precise and rapid determination of these shifts in the following way.

The rf oscillator was tuned to the point of maximum slope of the resonance. As the electric field was applied, the oscillator was simultaneously retuned to within 10Hz of the shifted point of maximum slope. The signal intensity changed by an amount proportional to the difference between the Stark shift and the change in the applied rf frequency. The constant of proportionality is simply the slope of the resonance curve, $\partial I/\partial f$, which is a constant to 1% within 10 Hz of the point of maximum slope. This constant was obtained by measuring the signal change produced by a 2-Hz shift of the oscillation frequency. Measurements were made on opposite sides of the resonance to subtract out resonant-independent modulation of the observed signal. Electric field measurements of both polarities were averaged

together to eliminate linear electric field dependencies due to instrumental ($v \times E$) effect.[17]

At each field point the rf amplitude was varied from the minimum at which a discernable resonance appears up to 1 V peak-to-peak, or until the shape of the rf dependence curve was established (see Fig. 1). This rf dependence arises from the multiple quantum transitions[18] induced in the five-level system. The flop-in transition is isolated from the others only at zero rf power: hence, we must extrapolate the rf dependence curves to zero to obtain the Stark shift of these levels.

The observed shift multiplied by the ratio of the loop separation to the length of the electric field plates (filling factor) is the true shift arising from the tensor polarizability.

IV. APPARATUS

A complete description of the apparatus can be found in Ref. 17. Gallium beams were produced in an oven made of boron nitride to prevent creep of the liquid up the walls. It was resistively heated by wires of tantalum.

The beam was detected on a tungsten oxide surface prepared by reducing tungsten carbonyl using a technique similar to that of Greene.[19]

The rf fields generated by a General Radio 1163A coherent decade synthesizer were stable to one part in 10^8 per day. The loops made of 1-cm-wide copper strips were suited for radio frequency transitions. Producing the 2-GHz fields necessary for a study of the $\Delta F = 1$ hyperfine transition, which would have shed additional light on the nature of the polarizability, was beyond the range of this equipment.

The glass electric field plates have been described by Gould.[20] The plates are 1.90-cm-thick by 6.35-cm-wide by 71.12-cm-long solda-lime glass, which becomes electrically conducting when heated above 120 °C.[21,22] The plates were conditioned to sustain fields up to 450 kV/cm.[20,23] The spacing of the plates was measured to be 0.1416 ± 0.0013 cm., and this produced the limiting uncertainty in our results.

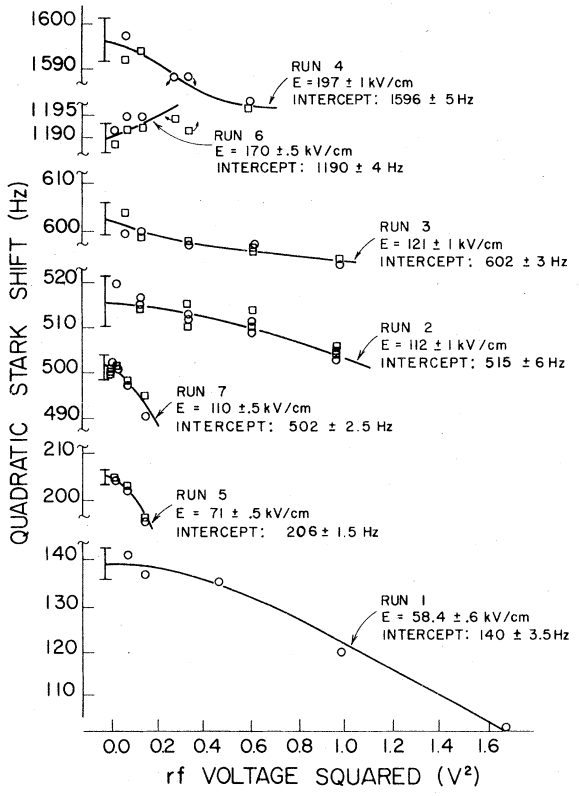

FIG. 1. Experimental values of quadratic Stark shift of the "flop-in" transition of gallium. Points are plotted vs the square of the rf voltage inducing the transition for seven different values of the electric field. Open circles: points taken with the electric field parallel to the magnetic axis; open boxes: points with the field antiparallel. For each value of E the observed shift is extrapolated to its value with zero rf power.

V. RESULTS

The quadratic Stark shift of the flop-in transition of a beam of naturally occurring gallium was measured at seven electric fields. The excitation of multiple-quantum transitions was considerable, as seen from the rf power dependence curves of Fig. 1. The behavior of the multiple-quantum transitions observed at rf voltages approaching zero agrees with the description given in Salwen,[24] Pegg,[25] and Kusch.[26] At low rf power, only the single- and double-quantum transitions will be significant, and the Stark-shift dependence on the square of the rf voltage flattens out near zero. The scatter in the several runs at each field strength leads to an uncertainty in the extrapolated value of less than $\frac{1}{2}\%$.

The seven points plus the origin are plotted against the square of the field strength (Fig. 2), and a least-squares fit shows only a 0.07% standard deviation. The slope of this line corrected for the filling factor (1.20) and a small asymmetry factor[27] is $K_2 = 500.1 \times 10^{-10}$ Hz/(V/cm)^2.

Uncertainties in the filling factor are 1.2%. The plate separation and linearity of the plate high-voltage dividing network each contribute 2% uncertainty to the field strength squared. Extrapolations and other effects are determined to within 0.6%.

The tensor polarizability from Eq. (2) is

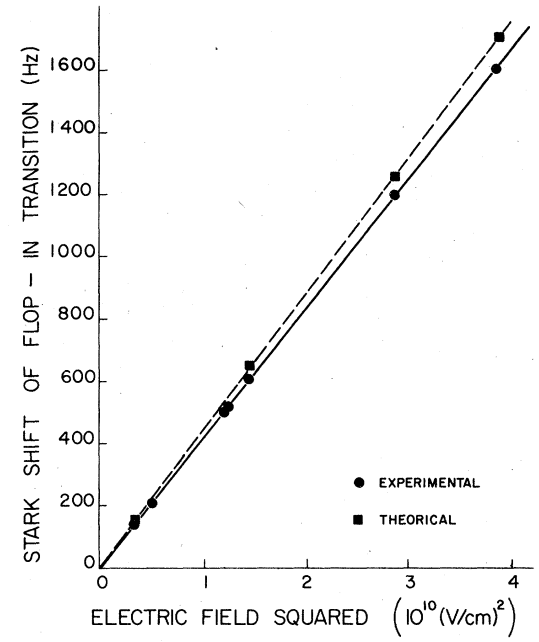

FIG. 2. Solid circle: plot of the zero rf power extrapolated Stark shift as a function of the square of the magnitude of the electric field applied including the origin. These points lie on a straight line whose slope is 410.9 (0.3) $\times 10^{-10}$ Hz/(V/cm)^2. Solid boxes: plot of theoretical values of transiton frequency at various electric fields minus the frequency at $E=0$ as measured from the computer generated net transition probabilities. These points lie on a straight line whose slope is 440.2(0.5) $\times 10^{-10}$ Hz/(V/cm)^2.

$$\alpha_t = \tfrac{4}{3}K_2, \tag{15}$$

or

$$\alpha_t = 667(20)\times 10^{-10}\ \text{Hz/(V/cm)}^2. \tag{16}$$

VI. COMPARISON

Theoretical transition frequencies were calculated for each isotope at various fields to second order in the Breit-Rabi formula using the polarizabilities from Eqs. (13) and (14). A Ramsey pattern characteristic of the machine used was drawn for each isotope centered at the transition frequency and weighted by the isotopic abundance, 60.1:39.9, by a PDP-10 computer with graphic facilities. The computer also plotted the sum of the patterns at each field strength to represent the actual resonance to be expected in our experiment. Some of these plots are shown in Fig. 3. By fitting an interval equal to the experimental linewidth, 380 Hz, between points of equal intensity within the central valley we determined the transition frequency and, subtracting the zero electric field frequency, we compute the apparent Stark shift.

These values are plotted in Fig. 2 and lie in a straight line defining an effective polarizability $\alpha = 714(16)\times 10^{-10}$ Hz/(V/cm)^2, the uncertainty here arises mostly from the uncertainty in the values used for α_t of the $J=\frac{3}{2}$ state. We note that for all fields within the range observed the com-

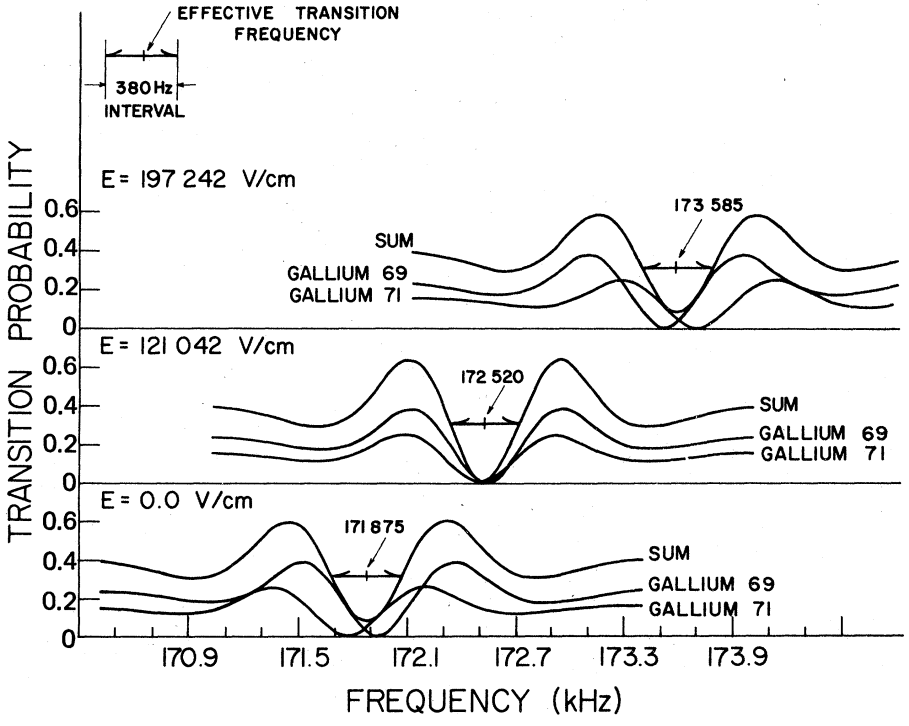

FIG. 3. Computer-generated Ramsey patterns for the flop-in transition of the two gallium isotopes at various electric field strengths; above is the abundance weighted superimposed pattern. The magnetic field is 0.740 G.

puter simulated measurements predict an effective polarizability very close to the weighted average of the individual theoretical polarizabilities of the two isotopes. This is because, in this range, the difference in the shift of these isotopes is never greater than half the linewidth of the resonance, hence significant interference in the summed pattern does not occur; this would cause an asymmetry which could significantly change the effective polarizability.

In both the experiment and the theoretical model the shift of the resonance is linear in the square of the electric field strength, and the line shape is not discernibly changed. The values for the polarizability agree within the limits of error.

Angel, Sandars, and Woodgate[9] used similar calculations on the ground state of aluminum and found a theoretical value 14% higher than that which they measured. The present work demonstrates an agreement, to within 7% of a measured atomic polarizability with theoretical results. The special nature of the stark effect in a $^2P_{1/2}$ atomic state is thus confirmed experimentally.

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[1]R. D. Haun and J. R. Zacharias, Phys. Rev. 107, 107 (1957).
[2]E. Lipworth and P. G. H. Sandars, Phys. Rev. Lett. 13, 718 (1964).
[3]H. Gould, E. Lipworth, and M. C. Weisskopf, Phys. Rev. 188, 24 (1969).
[4]J. P. Carrico, A. Adler, M. R. Baker, S. Legowski, E. Lipworth, P. G. H. Sandars, T. S. Stein, and M. C. Weisskopf, Phys. Rev. 170, 64 (1968).
[5]J. R. Mowat, Phys. Rev. A 5, 1059 (1972).
[6]P. G. H. Sandars, Proc. Phys. Soc. London 92, 857 (1967).
[7]J. R. P. Angel and P. G. H. Sandars, Proc. R. Soc. London Ser. A 305, 125 (1968).
[8]A. Khadjavi, A. Lurio, and W. Happer, Phys. Rev. 167, 128 (1968).
[9]J. R. P. Angel, P. G. H. Sandars, and G. K. Woodgate, Proc. R. Soc. London Ser. A 388, 95 (1974).
[10]R. T. Daly and J. H. Holloway, Phys. Rev. Lett. 96, 539 (1954).
[11]A. Lurio and A. G. Prodell, Phys. Rev. 101, 79 (1956).
[12]C. Schwartz, Phys. Rev. 97, 380 (1955).
[13]P. G. H. Sandars and J. Beck, Proc. R. Soc. London Ser. A 289, 97 (1965).
[14]C. Schwartz, Phys. Rev. 105, 173 (1957).
[15]R. F. Bacher and S. Goudsmit, *Atomic Energy States* (Greenwood, Westport, CT, 1978).
[16]F. R. Petersen, Bull. Am. Phys. Soc. 14, 833 (1969).
[17]S. J. Rosenthal, Ph.D. thesis (Brandeis University, 1974) (unpublished), University Microfilms.
[18]N. F. Ramsey, *Molecular Beams*, 2nd ed. (Oxford University P., London, 1963), p. 130.
[19]E. F. Greene, Rev. Sci. Instrum. 32, 860 (1961).
[20]H. Gould, Ph.D. thesis (Brandeis University, 1970) (unpublished), University Microfilms.
[21]R. Marrus, E. C. Wang, and J. Yellin, Phys. Rev. 177, 122 (1969).
[22]J. J. Murray, "Glass Cathodes in Vacuum Insulated High-Voltage Systems," University of California, U.C.R.L. Report No. 9506, 1960 (unpublished).
[23]G. P. Beukema, Physica (Utrecht) 61, 259 (1972).
[24]H. Salwen, Phys. Rev. 101, 621 (1956).
[25]D. T. Pegg, Phys. Rev. A 8, 2214 (1973).
[26]P. Kusch, Phys. Rev. 101, 627 (1956).
[27]Reference 18, p. 143.