Measurements of g Factors by a Beam-Foil Quantum-Beat Technique*

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The Landé g factors for some excited levels in ^{40}Ar II and III are measured by a beam-foil spectroscopic quantum-beat technique. The values are obtained from ratios of g factors, which are made with a standard g value taken as known. Where comparison is possible, the results of the measurements are generally in agreement with previous Zeeman measurements. In cases where discrepancies are found, the sources of the disagreement are discussed and identified. The technique may be applied to other elements having excited states with lifetimes in the nanosecond range. Other types of g-factor measurements for such states are not easily applicable.

I. INTRODUCTION

Beam-foil spectroscopy[1] is well established as a versatile technique for the observation and study of the spectra of atoms in various stages of ionization.[2] Previously unobserved transitions have been excited in a number of elements, and the mean lives of many excited states have been measured.[2] The beam-foil interaction also creates coherent alignment in those excited states which have electronic angular momentum $J > \frac{1}{2}$.[3] This alignment is assumed to arise from charge capture by the fast ion at the second surface of the foil, but the exact nature and characteristics of this process are currently a subject of investigation.

These properties of ionization, excitation, and alignment, characteristic of the beam-foil collision process, make possible the measurement of g factors of a number of levels not accessible to the usual method of Zeeman spectroscopy. The spectral lines observed in a discharge are often limited to the arc and first spark spectra. In particular, very few g factors for levels of the light elements in any stage of ionization have been measured.[4] Many of these states may be studied by the beam-foil method. Measurements of this type should aid in the identification of the atomic levels from which the observed spectral lines arise. Since some lines are only observed by the foil excitation, obtaining g factors from the same measurement assumes even more significance. Knowledge of the g factor of a level also permits a lifetime measurement by use of the Hanle effect.[3]

We discuss the measurement of g factors for excited levels of Ar II and III. These measurements represent an extension of the method initially applied to the first three charge states of neon.[5] The neon results were relative comparisons of the g factor of each level with a known g factor. This known g factor (of the $3p\,^3D_3$ level of Ne I) had been measured by Zeeman spectroscopy,[6] by the Hanle effect,[3] and by rf spectroscopy[3] with identical results in terms of accuracy. Although many precautions were taken to avoid systematic error, of our three neon g factor measurements which could be compared with the results of other methods, two were in disagreement outside the range of statistical uncertainty.[5] Among possible sources of error in our measurements are faulty line identification, blending of transitions, and transfer of alignment to the state under investigation through optical cascades from higher-energy aligned states. Because the g factors of many levels of Ar I and II have been measured, and because many of these levels are the sources of transitions in a convenient region of the spectrum, we chose this element to perform a series of calibration measurements. This choice also permitted the comparison of the alignment properties of foil collisions for equivalent charge states of elements with greatly different mass. The goal of our current measurements is to discover the range of applicability of this beam-foil method of g-factor measurement, to establish its accuracy, and to investigate sources of systematic error associated with its use. To this end we have measured the g values of excited levels of several charge states with small alignment, low intensity, blends, etc., as well as more favorable cases.

II. EXPERIMENTAL METHOD

The method of measurement has already been briefly presented,[5] but for convenience a short description is given here. Earlier measurements[3] have shown that m_J substates of a level characterized by the quantum numbers L, S, and J may be coherently aligned by the ion-foil collision. A uniform magnetic field H is applied perpendicular to the beam direction. When a given linear polarization component of the light is observed in a direction perpendicular to both beam and magnetic field, coherent precession of the angular momentum components about the field direction produces an intensity modulation of the light emitted from the level

in question, if alignment exists. This intensity modulation is extended in space by the rectilinear motion of the fast ion. However, in the measurements discussed here, a fixed point of observation d relative to the exciting foil is selected, and a sinusoidal modulation of the polarized light intensity is observed as a function of the linearly swept magnetic field strength.[3,5] The modulation of the intensity of light emitted by level k may be written

$$I_k(\omega_k, d) = A_k[1 + B_k \cos(2\omega_k d/v)]\, e^{-\Gamma_k d/v}, \qquad (1)$$

when there is no nuclear spin. Here Γ_k is the reciprocal mean life of the level, and the Larmor frequency is $\omega_k = 2\pi g_J(k)\mu_\beta H/h$. $A_k(1+B_k)$ is the total intensity at the foil, while B_k is the relative quantum-beat amplitude. A density-matrix analysis of an analogous experiment involving a coherently aligned excited state[7] shows that the magnitudes of the off-diagonal matrix elements are related to the relative alignment of the level. This general behavior is a consequence of the unitary transformation by which these off-diagonal elements are generated. The off-diagonal matrix elements provide a description of the coherence which is responsible for the quantum beats. This coherence is a consequence of the foil collision. The description of the quantum beats given by Eq. (1) shows that nonzero B_k implies both coherence and alignment. If there were no coherence, B_k would be zero whether alignment existed or not. If the level cannot be aligned ($J \leq \frac{1}{2}$), or if the cross sections for population of the m_J levels are equal, B_k is necessarily zero. From (1), $g_J = hv/(2\mu_\beta \overline{H} d)$, where $\overline{H}$ is the change in magnetic field strength required to produce a single period of intensity oscillation, μ_β is the Bohr magneton, h is Planck's constant, and v is the ion velocity.[5] The values of v, d, and $\overline{H}$ must be measured to about 1% in order to produce an accuracy of a few percent in an absolute measurement. Of the three, perhaps the velocity presents the greatest difficulty, since it is the velocity of the ions after the foil collision that is required. Not only must the accelerator terminal voltage be calibrated over the energy range of interest, but the beam-ion energy loss due to electronic and nuclear stopping during passage through the foil must also be accurately determined. In the absence of a post-foil velocity analyzer, this presents a formidable problem. An alternative approach to such an absolute measurement is to obtain the g values, ($g_J = g'\overline{H}'/\overline{H}_J$) in terms of a known g factor g' of the same element and the relative periods of the light-intensity oscillations in the same linear magnetic field sweep. In this case the velocity drops out, even for different charge states of the same element.[8]

III. APPARATUS

A beam of $^{40}Ar^+$ ions was accelerated by a 2-MV Van de Graaff generator to energies between 425 and 725 keV. Beam currents in the range 0.7–1.3 μA were passed through carbon foils with surface densities typically 10 $\mu g/cm^2$. The thickness of foils made in this laboratory was calibrated to 10% accuracy,[9] and initially ranged from 8.5 to 11.5 $\mu g/cm^2$ surface density. However, we found it important to use foils of nominally equal thickness during a given comparison measurement.[8] Our final measurements were made using 10-$\mu g/cm^2$ foils which varied essentially statistically in thickness. Several foils were used to collect each datum. No velocity corrections for different foil thickness were made, which probably resulted in small nonstatistical deviations in some of our early data.

Statistical fluctuations in the ion-beam current were not greater than ± 5%. On rare occasions, current variations with a long period were observed. When accidentally synchronized with the magnetic field sweep, spurious light-intensity variations resulted. Following the discovery of this possibility, the beam current was frequently checked for its existence, and the measurement cycle changed to better than average such variations.

Heavy ions, at the energies we use, rapidly destroy the thin unsupported carbon foils. We attempted to reduce this effect by using a foil backed with a mesh made of electrodeposited nickel.[10] This produced up to a factor of 2 longer foil life at energies below 500 keV, but essentially the same foil life and relatively less light at energies near 700 keV. The mesh withstood the ion bombardment relatively well, and new foils were repeatedly mounted. The increased lifetime of the foils was not maintained with this practice, and eventually the use of the mesh became a liability, even at the lowest energies. Results of the g-factor measurements were found to be independent of the presence of the mesh.

Light polarized parallel to the beam direction, emitted about the perpendicular to the ion beam from a 100-μ length of the beam at $d = 1$ cm,[3] was magnified a factor of 2 by a double-lens system to fill the grating of a Perkin-Elmer 210B monochromator. Photon-generated pulses from a cooled EMI 6256s (S-13) photomultiplier tube were accumulated in a multiscaler as a function of the magnetic field strength applied perpendicular to both the beam and the direction of light collection. The magnetic field and the storage address of the multiscaler were stepped linearly with time.

The g-factor ratio measurements in neon had been made consecutively between each level and the standard, to ensure that the beam velocity was truly a constant.[5] Following each pair of measurements the ion beam had been switched off to

permit installation of new foils. The apparatus was modified for the argon measurements by adding a movable Faraday cup and a valve before the foil wheel. This permitted continuous stabilization of the ion beam for a whole sequence of measurements, during which the measurement chamber could be isolated. The control of the beam energy was such as to maintain the velocity of the ions constant to 0.1% during a measurement sequence.

IV. RESULTS AND DISCUSSION

We chose to investigate the strongest argon spectral lines in the interval 3000–5000 Å which appeared to be unblended, using moderate spectrometer resolution. The point of observation was 1 cm from the foil. More intense light was observed at lower beam energies in nearly all cases. To investigate line shapes, a single lens, well separated from the beam, focused the light originating approximately 1 cm from the foil onto the spectrometer entrance slit which was 100 μ wide. Single lines then had an observed full width at half-maximum of 4–5 Å. Blends of the transitions near 3543, 3560, 4346, and 4423 Å were clearly observed as relatively broader lines. The remaining lines which we chose to investigate appeared to be single lines, although the weaker lines could not be studied under these conditions. Most of the observed lines were clearly strong lines of the Ar II and Ar III spectra, with only the strongest lines of Ar I possibly appearing.[11] Our operational definition of lack of blending was to observe a pronounced intensity dip between lines in a spectroscopic measurement made on lines thought to be single under the conditions used for quantum-beat measurements. With the spectrometer slits widened to 200 μ for measurements, an apparent resolution meeting this criterion was found for all transitions investigated, except for those previously identified as blends. However, subsequent measurements of another type, discussed below, showed that a slight amount of blending probably existed for some "unblended" transitions.

The spectral line near 4610 Å originating from the Ar II level $4p'^2F^\circ_{7/2}$, with $g = 1.140$ from Zeeman measurements,[4] was intense and relatively highly aligned. This level was chosen as the standard, in terms of which the g factors of the other levels were measured. We have not yet made an absolute measurement of this g value, but a departure of our measurements from the Zeeman value would be indicated by a systematic deviation in the g values measured for all other levels. No such systematic deviation was found.

Table I shows the results of the g-factor measurements for lines where little blending was observed. The results of Zeeman measurements[4] are also presented. The agreement with the Zeeman measurements is gratifying, but the statistical uncertainties of single measurements (typically ± 0.05) are rather large in comparison both with those possible in the neon work,[5] and with those of the Zeeman effect. This is due to the relatively smaller alignment in argon, and to the shorter foil lifetime mentioned earlier, which made collection of extensive data difficult. However, some measurements have larger fluctuations than other comparable ones. This is thought to be due to variations in energy loss of the beam ions, due to less careful foil-thickness selection in some of the early measurements.

The oscillation periods were obtained by a least-squares fit of the raw data to a function representing a constant background plus a sinusoid with adjustable phase. The frequency providing the best fit was chosen. Typically two full oscillations were present in the data for magnetic fields with magnitude less than 100 G. Motional Stark effects may be neglected in these weak fields except for those elements such as the lightest ones with unusually small configuration separation.[3] The statistical uncertainties quoted in the tables describe the standard deviation of the mean. From 3 to 11 in-

TABLE I. Results of g-value measurements on unblended transitions or transitions with minor blends. (Standard deviations of the uncertainty of the averages are given.)

Charge[a] state	Designation[a]	Wavelength[a] (Å)	Relative quantum-beat amplitude (%)	g value (our measurement)	g value (Zeeman measurement)[b]
. . .	. . .	4628	3.9 ± 0.45	1.092 ± 0.01	. . .
II	$4p\,^2D^\circ_{5/2}$	4879.86	3.4 ± 0.1	1.264 ± 0.022	1.241
II	$4p\,^2P^\circ_{3/2}$	4764.86	3.2 ± 0.25	1.205 ± 0.018	1.244
II	$4p'\,^2F^\circ_{7/2}$	4609.56	6.5 ± 0.2	taken as standard	1.140
II	$4p'\,^2F^\circ_{5/2}$	4589.90	4.8 ± 0.8	0.886 ± 0.023	0.857
. . .	. . .	4579	3.8 ± 0.26	1.024 ± 0.022	. . .
III	$4p'\,^3F_4$	3336.13	2.2 ± 0.15	1.182 ± 0.025	. . .

[a]See Ref. 11.

[b]See Ref. 4.

dependent measurements were combined with equal weights. The reference g value was assumed to be known exactly. In three instances individual measurements deviated by four or more standard deviations from the remainder. These were not included in the averages. Ion-beam-current variation during the measurement may be the cause of these anomalies.

The relative quantum-beat amplitudes listed in Tables I and II were obtained by first subtracting the small photocount background from the data. The difference between the intensity maximum and minimum of the beat was then divided by twice the average intensity,[3] and the results averaged over a number of independent measurements. Relative quantum-beat amplitudes of 5–15% had been observed earlier for many states of the ions of neon, while Table I shows that analogous values for argon are typically a factor of 2 smaller. Ions in lower charge states seem to have greater alignment for a given J value. A slight tendency toward larger-amplitude quantum beats for excited states with large angular momentum may be seen.

Several transitions originating from Ar III states were examined. Only the $4p'\,^3F_4$ state had sufficient alignment and intensity to permit a measurement of even modest precision. The Landé formula yields $g = 1.250$ for this state. Our result indicates a deviation from pure LS coupling.

The levels $4p\,^2D^\circ_{5/2}$ and $4p\,^2P^\circ_{3/2}$ of Ar II have essentially the same g factors from Zeeman measurements. A direct ratio taken by our comparison method yielded a value 1.00 ± 0.025.

We originally identified the transition near 4628 Å as the strong transition of Ar I at this wavelength arising from the $5p\ [2\frac{1}{2}]_2$ level,[11] with $g = 1.09$ in excellent agreement with our result. In connection with another experiment, we subsequently obtained the mean life by the BFS method,[2] using a standard apparatus of this laboratory, and found it to be near 15 nsec; substantially shorter than the mean life of the Ar I level obtained by another method.[12] Hence, we leave this level unidentified. The BFS measurement also showed a probable blend with mean life near 2 nsec, which contributed less than 15% of the total light at $d = 1$ cm from the foil, and hence was not detected with the spectral scan.

We also found a short-lived blend which contributed about 5% of the light to the transition near 4880 Å, while a weak blend of the transition near 4590 Å is also possible. The evidence for the latter is ambiguous, however, as the appearance of a weak cascade forbade an exact interpretation of the decay. A cascade with mean life near 2 nsec to the level $4p\,^2P^\circ_{3/2}$ was definitely identified. The transitions near 4610 and 3336 Å showed no apparent blends or cascades.

It is difficult to associate the transition at 4579 Å with any known levels in Ar I–IV other than the $4p\,^2S_{1/2}$ level of Ar II. That level is the origin of a transition at 4579.35 Å which has an appropriate intensity, but such a level cannot be aligned. We observed quantum beats at several beam energies. The level identification is supported by Zeeman-effect measurements on this transition. The g value obtained[4] (1.695) does not agree with our result. We can only conclude that we have observed another, unidentified, transition. For a further check we investigated an analogous transition at 3510 Å, which has as a source two possible states, neither permitting alignment. No indication of quantum beats was observed for this transition.

A subsequent analysis of the BFS decay curve showed a two-time constant decay, with one-time constant near that of the mean life of the $4p\,^2S_{1/2}$ level.[13] We interpret this as a blend of two transitions, only one of which is polarized. In this case, a blend cannot affect the quantum beats, since the light from the nonaligned level is independent of magnetic field. A further discussion of this analysis is presented in another publication.[14]

The transition at 4805 Å gave clear evidence of alignment, but the beat pattern was weak and nonsinusoidal. In this case, it is not apparent that blending with another transition occurs. The partial transfer of coherence in a cascade process[15] funneled through this $4p\,^4P^\circ_{5/2}$ state may explain the observation. A number of transitions into this state have been identified.[11] Evidence of long-lived cascades was found with the beam-foil lifetime measurement, but in the absence of complementary measurements, the interpretation remains open. Only the two other transitions discussed above show evidence of cascades. These cases do not give evidence of any major effects, based on the g-value results. However, our measurements do not represent a sample sufficient to regard possible effects of cascades as negligible. The results of measurements on transitions where strong blending was apparent are exhibited in Table II. These measurements are included to show that the presence of relatively large-amplitude sinusoidal quantum beats is not sufficient to guarantee accurate results. The observed g values do not correspond to the g factors of any of the possible states, although the apparent deviations are in some cases small. It is easy to see that superposition of the light from different transitions may lead to an apparent frequency shift toward the mean value. This is apparently what occurs in these cases.

In certain instances one might imagine separating blended lines on the basis of different lifetime by appropriate choice of the parameter d, the observation distance from the foil. This was actually the case for the transitions near 4628 and 4880 Å. Selecting the appropriate beam energy

TABLE II. Results of g-value measurements on transitions with substantial or probable blends. (Standard deviations of the uncertainty of the averages are given.)

Charge[a] state	Designation[a]	Wavelength[a] (Å)	Relative quantum-beat amplitude (%)	g value (our measurement)	g value (Zeeman measurement)[b]
I	$5p[1\frac{1}{2}]_1$	4424.0			1.01
II	$4p^4P^o_{5/2}$	4420.9	2.2 ± 0.2	1.293 ± 0.015	1.720
II	$4p^4D^o_{5/2}$	4426.0			1.334
I	$5p'[1\frac{1}{2}]_1$	4345.17	2.7 ± 0.1	1.378 ± 0.012	0.61
II	$4p^4D^o_{7/2}$	4348.1			1.427
II	$4d'^2G_{7/2}$	3562.19			. . .
II	$4d'^2G_{9/2}$	3561.03	4.6 ± 0.13	1.079 ± 0.01	. . .
II	$4d^2F_{7/2}$	3559.51			1.167
II	$5d^2F_{7/2}$	3543.15	5.3[c]	0.947 ± 0.05	. . .
II	$4d^2F_{5/2}$	3545.60			0.861
II	$4d^4D_{1/2}$	3509.78	0 ± 0.4	not aligned	0.380
III	$4p^3P_0$	3509.33			. . .

[a]Identified transitions which may contribute to the observed blend, see Ref. 11.

[b]See Ref. 4.

[c]Single measurement.

could enhance lines from one charge state at the expense of another. In any case, the identification of blends appears to be an important factor in the determination of accurate g values by the method discussed here. An analysis of each instance of blending in terms of the observed relative intensities and alignments of the superposed transitions should be considered when measurements of this nature are necessary. A blend with a transition from a nonaligned state will not affect the g value obtained. Some of our "unblended" transitions turned out to have up to 15% of the total intensity arising from another transition, which could be partially polarized; but it is not apparent that this affected the g values. We estimate that the "blended" transitions had at least 70% superposition of light from other transitions, i.e., the transitions appeared to be single lines when observed with only moderate resolution. This type of blending produced g-value deviations of varying magnitude. No generally definitive rule for analysis was obtained from observation of these particular cases.

V. SUMMARY

We have measured the g factors of several levels of Ar II and III by a beam-foil spectroscopic quantum-beat technique. For those levels emitting spectral lines which were not blended, or with the light intensity contributed by the blend $\lesssim 15\%$, the results of the g-factor measurements are in good agreement with previous Zeeman observations. Discrepancies of various magnitudes were observed when transitions with substantial blending were investigated. Measurement of g factors with relative quantum-beat amplitudes as small as 2% was possible. Measurements of this type should aid in the identification of transitions in beam-foil spectra.

Because of their small alignment and the rapid destruction of foils at low energies, the excited levels of argon represent a poor choice for precision measurements. The fluctuations observed in individual measurements are attributed to the difficulty in obtaining sufficient statistics for averaging random fluctuations, and to use of unequal foil thickness for some comparison measurements. Choice of ions with smaller mass and larger relative alignment should permit measurements of substantially higher precision in the future. Work in progress toward this goal includes planned improvements in technique, and further study of the earlier g-factor measurements in neon.

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Hyperfine Structure of Excited States and Quadrupole Moment of Ne^{21} Using Laser-Induced Line-Narrowing Techniques*

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Optical hyperfine structure in Ne^{21} has been observed using laser-induced line-narrowing techniques. The output from a long stabilized single-mode 1.15-μ He-Ne laser is focused into an external sample cell containing Ne^{21}. The laser field resonates with the Ne^{21} 1.15-μ transition causing changes in the level populations over a narrow velocity range. Viewed along the laser axis, these population changes appear as narrow "change signals" superposed on the Doppler background of the spontaneous emission of the coupled 6096-Å transition. Analysis of the spectrum of these change signals, emitted parallel and antiparallel to the laser-field propagation direction, gives values for the hyperfine A constants for the three levels involved. These constants are analyzed by means of theoretical expressions wherein $\langle 1/r^3\rangle$ of the $2p$ hole is the single important parameter. Combining the resulting value of $\langle 1/r^3\rangle_{2p\,\mathrm{hole}}$ for the $1s_4$ level with a previous measurement of $B(1s_5)$ gives a value of $Q(1s) = (0.093 \pm 0.002)$ b, in agreement with an earlier less-accurate value. In addition, the $Ne^{20,21,22}$ isotope shifts are measured at 1.15 μ and 6096 Å.

INTRODUCTION

Laser-induced line-narrowing spectroscopy[1] has been used to observe optical hyperfine structure in Ne^{21}. This technique represents an advance in several ways over other methods of investigating hyperfine and other closely spaced structure in Doppler-broadened systems. The best conventional optical spectrometers, even if not instrument limited in resolution, are unable to resolve hyperfine transitions whose structure is buried within the Doppler profile. Atomic-beam studies in rare gases eliminate the Doppler effect, but have been generally limited to metastable levels, the only states with hyperfine structure that have lifetimes sufficiently long for beam techniques. Furthermore, ordinary beam techniques are incapable of measuring isotope shifts.

In the present approach, an optical technique[2-5] is used in which the Doppler width is effectively eliminated by laser-induced velocity selection. Measurements of the narrowed spectral lines have yielded values for the isotope shifts of $Ne^{20,21,22}$ at 1.15 μ and 6096 Å and hyperfine constants of Ne^{21}, which lead to an accurate value for the Ne^{21} quadrupole moment.

The level scheme under study (Fig. 1) consists of the hyperfine sublevels of the $2s_2$, $2p_4$, and $1s_4$ fine-structure levels[6] of Ne^{21} (nuclear spin $I = \frac{3}{2}$), which form the 1.15-μ ($2s_2 - 2p_4$) and 6096-Å ($2p_4 - 1s_4$) cascade transitions. The magnetic dipole and electric quadrupole hyperfine interactions split each fine-structure level (angular momentum J) into a number of hyperfine components, of energy