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g Factors of the $6s5d$ Configuration of Atomic Barium*

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The four metastable states of atomic barium comprising the $6s5d$ configuration have been studied by atomic-beam magnetic resonance, and information concerning their g factors has been obtained. Data from this work agree with that obtained recently by v. Oppen using a different method, and agreement is also obtained with theory.

INTRODUCTION

Atomic barium has a $^1S_0$ electronic ground state, and it is therefore necessary to make hyperfine-structure measurements in excited electronic states. The hyperfine structure of Ba^{137} has been studied in the $^1P_1$ state by level crossing,[1] and it would be worthwhile to extend this work to measurements in metastable electronic states. For this reason it was decided to attempt to observe the four metastable states of atomic barium belonging to the $6s5d$ configuration by use of the technique of atomic-beam magnetic resonance. These states have lifetimes of the order of seconds, and they should therefore be observable in a beam.[2]

The g factors of these metastable states have recently been measured by v. Oppen[3] by resonance scattering of light from an atomic beam of barium which had been partially aligned by both optical pumping and electron bombardment. Our results are in general agreement with his although the observational technique used was different, and our results therefore serve to confirm the values of the g factors for these states.

APPARATUS

The atomic-beam apparatus used in this work operated in the flop-in mode, and is of similar design to those used at Princeton having a 6-pole A magnet and a 2-pole B magnet. These are permanent magnets, and are located inside the vacuum envelope, while the C magnet is an electromagnet and is located outside of the vacuum. The transitions were excited by a simple radio-frequency loop which is located in the C-field region, and which obtains its rf power from a frequency synthesizer.

Since the excitation energy of these metastable states is of the order of 1 eV, it is not possible to detect a beam of them by secondary electron ejection from a metal surface. The beam was detected by electron-bombardment ionization followed by mass analysis by means of a detector which has been previously described.[4] The signal from this detector was averaged digitally, allowing the use of a fast, repetitive sweep so that the effect of fluctuations in beam intensity on the data could be minimized.

An atomic beam of barium is easily produced by heating a tantalum oven containing the metal. As it turned out, if this heating is accomplished by electron bombardment of the slit area of the oven, a sizable fraction of the beam is excited into the metastable states of interest. It was therefore possible to produce the beam and excite it in the same operation, thus simplifying the operation of the apparatus. This does not seem to be the most satisfactory method of opera-

tion, however, and in future work an attempt will be made to separate the functions of heating and excitation. Nevertheless, it was possible to obtain signal-to-noise rations of the order of 10 to 1 easily by this technique, and resonances were observed in all four of the metastable states.

RESULTS

Resonances were first observed in the $^1D_2$, $^3D_3$, and $^3D_2$ states of barium at magnetic fields of approximately 11.2 and 145.7 G, and the results are shown in Table I. The resonance linewidth observed in this work, which varied somewhat with applied magnetic field, averaged about 0.150 MHz. Since the magnetic field was not precisely known in these measurements the data are used only to determine the ratios of the g factors which should be the same as the ratios of the frequencies at these values of magnetic field. The experimental ratios can be compared with the Landé g factor ratios which are

$$g(^3D_3)/g(^1D_2) = 1.3341 \ ,$$
$$g(^3D_2)/g(^1D_2) = 1.1671 \ .$$

The corresponding experimental ratios as given by v. Oppen[3] are, respectively, 1.3299(5) and 1.1602(5). The experimental results are seen to be in agreement, and to differ slightly from ratios as determined from the Landé g factors.

The g factor of the $^3D_1$ state was measured absolutely by comparison of its resonance frequency with that of Cs^{133} at a field of approximately 450 G. Six measurements were made, and the resulting mean value is

$$g(^3D_1) = 0.498\,51(8) \ .$$

This value can be compared with v. Oppen's result of 0.4986(2) and with the Landé g factor, 0.498 84, and the experimental results can be seen to be in agreement while there appears to be a slight disagreement with the Landé g factor. Corrections in the g factor due to the nonlinear Zeeman effect at 450 G have been calculated, and are less than 2 in the fifth significant figure. The g factors of the other states were not measured absolutely, but the values are given by v. Oppen.[3]

DISCUSSION

The g factors of the $6s5d$ configuration of atomic barium have now been measured by two different experimental techniques and the results are in agreement. The errors which we quote represent about twice the spread in the data, and no attempt was made to analyze the data in a statistically meaningful manner. Furthermore, this apparatus has not been used sufficiently for measurements of this type to allow an evaluation of possible systematic errors in our value of the g factor of the $^3D_1$ state.

Our g-factor ratios differ from the Landé ratios. We have fit the experimental fine-structure separations to theory, and have obtained $G_2 = 990.2\ \mathrm{cm}^{-1}$, $\zeta = 237.0\ \mathrm{cm}^{-1}$, and $\eta = -11.93\ \mathrm{cm}^{-1}$, where G_2, ζ, and η are the electrostatic, spin-orbit, and spin–other-orbit parameters.[5] With three parameters and three energy differences we can reproduce the experimental fine-structure separations exactly. From these values we obtain, for the wave functions in LS coupling,

$$\psi(^3D_2') = a\psi(^3D_2) - b\psi(^1D_2)$$
$$= 0.989\,97\psi(^3D_2) - 0.141\,27\psi(^1D_2) \ ,$$
$$\psi(^1D_2') = b\psi(^3D_2) + a\psi(^1D_2)$$
$$= 0.141\,27\psi(^3D_2) + 0.989\,97\psi(^1D_2) \ .$$

The g factors in intermediate coupling are

$$g(^3D_2') = a^2 g(^3D_2) + b^2 g(^1D_2) = 1.1638 \ ,$$
$$g(^1D_2') = b^2 g(^3D_2) + a^2 g(^1D_2) = 1.0033 \ ,$$

with $g(^3D_2) = 1.1671$ and $g(^1D_2) = 1.0000$. These values are in agreement with those calculated by v. Oppen. The ratios of the theoretical g factors are then

$$g(^3D_3)/g(^1D_2') = 1.3297 \ ,$$
$$g(^3D_2')/g(^1D_2') = 1.1599 \ .$$

These ratios are in much better agreement with experiment than ratios of the Landé g factors as is to be expected. If spin-other orbit corrections are ignored, we obtain $g(^3D_2') = 1.1644$ and $g(^1D_2') = 1.0027$. The ratios of the g factors are then slightly high, but within experimental error.

TABLE I. Resonances used to determine g-factor ratios at magnetic fields of 11.2 and 145.7 G.

Magnetic field, G	$\nu(^1D_2)$, MHz	$\nu(^3D_3)$, MHz	$\nu(^3D_2)$, MHz	$\frac{\nu(^3D_3)}{\nu(^1D_2)}$	$\frac{\nu(^3D_2)}{\nu(^1D_2)}$
11.2	15.635(4)	20.806(8)	18.145(4)	1.3307(9)	1.1605(6)
145.7	203.613(40)	270.808(40)	236.265(40)	1.3300(6)	1.1603(6)
			Mean	1.3303(11)	1.1604(9)

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Pauli-Principle Restriction on the Two Matrix of Bopp for Atomic Ground States*

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A lower bound to the ground-state energy of an atom is formulated, using the pair density matrix $\Gamma^{(2)}$. The major problem in the development of such methods is that of finding necessary conditions on $\Gamma^{(2)}$ not satisfied by the current optimal class of density matrices. It is shown that the Bopp two-matrix ansatz suffers mainly from the Pauli principle not being satisfied. With the aid of an extensive set of system-dependent identities for $\Gamma^{(2)}$, numerical computations indicate how energy improvement will follow from insistence on the Pauli principle. A new necessary condition is conjectured which combines the Pauli condition on $\Gamma^{(1)}$ with a maximum eigenvalue condition on $\Gamma^{(2)}$.

1. INTRODUCTION

Quantum-mechanical calculations for systems of more than two particles are beset by the enormous difficulties of working with wave functions of many variables. In order to avoid the use of a large number of coordinates in obtaining energies – and to some extent, expectation values – studies have been made[1] of the possibility of using reduced two-particle density matrices alone in such computations. Instead of working with an N-particle wave function $\psi(x_1, \ldots, x_N)$ to find the ground-state properties of the (identical particle) Hamiltonian

$$H = \sum_{i=1}^{N} T(i) + \frac{1}{2} \sum_{i \neq j = 1}^{N} v(i,j), \tag{1.1}$$

one writes the Rayleigh-Ritz principle directly in terms of the two-body density matrix:

$$E_0 = \min_{f^{(2)}} \tfrac{1}{2} N \operatorname{Tr}(H^{\mathrm{red}} f^{(2)}),$$

where

$$H^{\mathrm{red}} \equiv T(1) + T(2) + (N-1)v(1,2),$$

and

$$f^{(2)}(1'2'|1\,2) \equiv \sum_\alpha C_\alpha \int \psi_\alpha(x_1' x_2' x_3 \cdots x_N) \times \psi_\alpha^*(x_1 x_2 x_3 \cdots x_N)\, dx_3 \cdots dx_N, \tag{1.2}$$

with $\sum_\alpha C_\alpha = 1$, $C_\alpha \geq 0$.

The idea is to vary over $f^{(2)}$, and the difficulty is