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Cascade Anticrossing Measurement of the Anomalous Hyperfine Structure of the 4^2D State of Rubidium*

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A new technique, cascade anticrossing spectroscopy, has been developed to determine the effective values of $\langle r^{-3}\rangle$ for the spin-dipole, orbital, contact, and quadrupole hyperfine interaction terms for the 4^2D state of Rb. They are, in units of 10^{24} cm^{-3}, $\langle r^{-3}\rangle_d = -0.36(8)$, $\langle r^{-3}\rangle_l = +0.20(2)$, $\langle r^{-3}\rangle_c = -4.3(1)$, and $\langle r^{-3}\rangle_q = +0.79(66)$, respectively. The negative value of $\langle r^{-3}\rangle_d$ and the unexpectedly large value of $\langle r^{-3}\rangle_c$ are surprising. The fine-structure splitting has been measured to be $-13\,360.9(8)$ MHz.

Recently, experimental investigations of the D-state hyperfine structures of alkali atoms[1] have revealed that many $D_{5/2}$ states have anomalous negative magnetic-dipole coupling constants. In order to gain a better understanding of these anomalies we have used a new spectroscopic technique, cascade anticrossing spectroscopy, to determine the complete hyperfine Hamiltonian for the 4^2D state of rubidium. The results of this work are quite surprising. The core polarization of the $4D$ state is substantially larger than the core polarization of any of the P states of rubidium, and the effective value of $\langle r^{-3}\rangle$ for the dipole contribution to the hyperfine structure is *negative*. Although many instances are known[2] where the effective values of $\langle r^{-3}\rangle$ for the dipole and orbital contributions to the hyperfine structure are not the same, the differences seldom exceed 10%, and we are not aware of any other report of a negative value for $\langle r^{-3}\rangle$.

The magnetic dipole and electric quadrupole contribution to the hyperfine-structure Hamiltonian of a hydrogenic atom can be written as

$$\mathcal{H}_{\rm hfs} = \mathcal{H}_l + \mathcal{H}_d + \mathcal{H}_c + \mathcal{H}_q . \tag{1}$$

The electric quadrupole contribution is

$$\mathcal{H}_q = \frac{2LQe^2}{2L+3}\langle r^{-3}\rangle_q \frac{3(\vec{\mathbf{L}}\cdot\vec{\mathbf{I}})^2 + \frac{3}{2}(\vec{\mathbf{L}}\cdot\vec{\mathbf{I}}) - L(L+1)I(I+1)}{2L(2L-1)I(2I-1)}, \tag{2}$$

where Q is the nuclear quadrupole moment. The orbital contribution is

$$\mathcal{H} = [(2\mu_B\mu_I)/I]\langle r^{-3}\rangle_l \vec{\mathbf{L}}\cdot\vec{\mathbf{I}} \tag{3}$$

and the dipole contribution is

$$\mathcal{H}_d = \frac{4\mu_B\mu_I}{I}\langle r^{-3}\rangle_d \frac{L(L+1)}{(2L-1)(2L+3)}\left[\vec{\mathbf{S}}\cdot\vec{\mathbf{I}} - 3\left\{\frac{\vec{\mathbf{S}}\cdot\vec{\mathbf{L}}\vec{\mathbf{L}}\cdot\vec{\mathbf{I}} + \vec{\mathbf{I}}\cdot\vec{\mathbf{L}}\vec{\mathbf{L}}\cdot\vec{\mathbf{S}}}{2L(L+1)}\right\}\right]. \tag{4}$$

The contact interaction is

$$\mathcal{H}_c = \frac{\mu_B \mu_I}{I} g_s \frac{8\pi}{3} |\Psi(0)|^2 \vec{S}\cdot\vec{I} \equiv \frac{2\mu_B \mu_I}{I} \langle r^{-3}\rangle_c \vec{S}\cdot\vec{I}. \quad (5)$$

For a nonrelativistic hydrogenic atom $\langle r^{-3}\rangle_q$, $\langle r^{-3}\rangle_l$, and $\langle r^{-3}\rangle_d$ are identical and are the inverse cube of the distance of the electron from the nucleus. The parameter $\langle r^{-3}\rangle_c$, which is defined by (5), is introduced for convenience of notation and is a measure of $|\Psi(0)|^2$ rather than the inverse cube of the distance of the electron from the nucleus.

We may also describe the hyperfine structure of the 4^2D state of rubidium with the Hamiltonian (1). However, since configuration interactions modify the hyperfine interactions, we must regard the quantities $\langle r^{-3}\rangle_i$ ($i = q$, l, d, and c) as convenient independent parameters which characterize the strengths of the interactions (2), (3), (4), and (5). The parameters $\langle r^{-3}\rangle_i$ ($i = q$, l, d, and c) do not necessarily represent, even approximately, the inverse cube of the distance of the valence electron from the nucleus.

The Hamiltonian which describes the fine structure and hyperfine structure of the 4^2D state in an external magnetic field H is therefore

$$\mathcal{H} = \mathcal{H}_{hfs} + h\xi \vec{L}\cdot\vec{S} + \mu_B H (g_l L_z + g_s S_z - g_I I_z). \quad (6)$$

We assume that $g_l = 1$ and $g_s = 2.002\,32$, and we use the nuclear g values tabulated by Fuller and Cohen[3] ($g_I = 0.999\times10^{-3}$ for Rb^{87}; $g_I = 0.295\times10^{-3}$ for Rb^{85}).

At low magnetic fields where $J = |\vec{L}+\vec{S}|$ is a good quantum number, the hfs Hamiltonian (1) reduces to

$$\mathcal{H}_{hfs} = hA_J \vec{J}\cdot\vec{I} + \mathcal{H}_q, \quad (7)$$

where, for $l = 2$ and $s = \frac{1}{2}$,

$$A_{3/2} = \frac{2\mu_B\mu_I}{hI}\left\{\tfrac{6}{5}\langle r^{-3}\rangle_l + \tfrac{2}{5}\langle r^{-3}\rangle_d - \tfrac{1}{5}\langle r^{-3}\rangle_c\right\}, \quad (8)$$

$$A_{5/2} = \frac{2\mu_B\mu_I}{hI}\left\{\tfrac{4}{5}\langle r^{-3}\rangle_l - \tfrac{4}{35}\langle r^{-3}\rangle_d + \tfrac{1}{5}\langle r^{-3}\rangle_c\right\}. \quad (9)$$

We have measured the low-field magnetic dipole coupling constants by cascade radiofrequency spectroscopy which are shown in Table I. The quadrupole interaction is too small to be measured in the low-field experiments.

Unfortunately the two measured values of $A_{3/2}$ and $A_{5/2}$ are insufficient to determine the three values of $\langle r^{-3}\rangle_i$ ($i = l$, d, and c). Additional information can be obtained by making measurements at high magnetic fields where J is not a good quantum number. The doublet splitting of the $4D$ state of rubidium was measured by Ramb[4] to be $\Delta\nu/c = -0.44\ \mathrm{cm}^{-1}$. Thus, a magnetic field of a few thousand gauss is adequate to decouple L and S. A diagram of the Zeeman splitting of the $4D$ state is shown in Fig. 1(b). As the Zeeman sublevels diverge from the zero-field dou-

TABLE I. Measured and least-squares fitted values of the magnetic dipole coupling constants A (MHz) and the positions of the anticrossing signals H (gauss). The values of A have been determined by cascade radiofrequency spectroscopy, while the values of H have been determined by cascade anticrossing spectroscopy.

Isotopes	Magnetic dipole coupling constants A or position of anticrossings $H(m_F)$	Measured values	Least-squares fitted values
Rb^{85}	$A_{3/2}$	7.3 (5)	7.26
	$A_{5/2}$	−5.2 (3)	−5.03
Rb^{87}	$A_{3/2}$	25.1 (9)	24.58
	$A_{5/2}$	−16.9 (6)	−17.03
Rb^{85}	$H(3)$	3815.18(24)	3815.38
	$H(2)$	3812.99(32)	3812.97
	$H(1)$	3810.29(32)	3810.23
	$H(0)$	3807.07(42)	3807.15
	$H(2)$	4790.31(42)	4790.00
	$H(1)$	4795.08(42)	4794.91
Rb^{87}	$H(2)$	3802.28(20)	3802.28

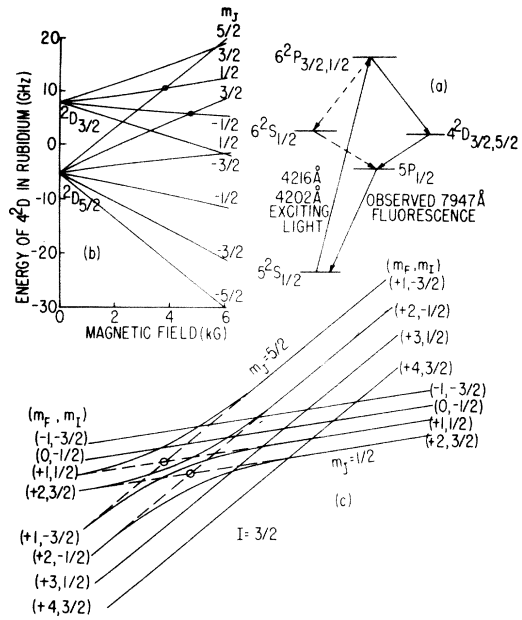

FIG. 1. (a) The energy level scheme of the relevant states in this work. (b) Zeeman splittings of the fine-structure levels in 4^2D state of Rb in a magnetic field. (c) The hyperfine levels involved at the 3800 G crossing in the 4^2D state of Rb^{87}. Circles indicate the positions of the anticrossings.

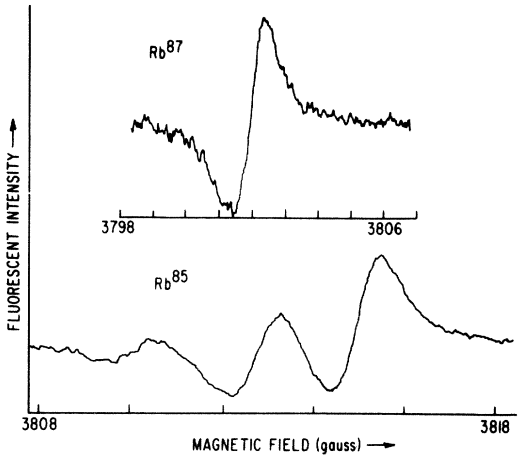

FIG. 2. Typical anticrossing signal recordings with σ exciting light. (Rb^{87} resonance line has been used in both cases.) The magnitudes of anticrossing signals depend, among other things, on the polarization and the line profile of the exciting light. Some signals are too small to be observed with σ excitation; however, they can be readily observed with π excitation.

blet a number of level crossings occur. If a nuclear spin is present each "crossing" consists of a large number of crossings and anticrossings, as shown in Fig. 1(c). The anticrossings are due to the hyperfine coupling of levels with the same values of the total azimuthal quantum number $m_F = m_I + m_J$.

In our cascade anticrossing experiments we excite the $6P$ state of rubidium with 4216- and 4202-Å resonance light from an electrodeless discharge lamp. We observe the intensity of the 7947-Å fluorescence from the $5P_{1/2}$ state, which is populated by cascades through the $6S$ and $4D$ states [Fig. 1(a)]. A consideration of the optical excitation rates and the branching ratios of the atom in a large magnetic field shows that in the neighborhood of the 3800-G $\Delta m_J = 2$ level crossing certain sublevels $|i\rangle = |m_J = \frac{5}{2}, m_I\rangle$ are more strongly populated than the sublevels $|j\rangle = |m_J = \frac{1}{2}, m_I + 2\rangle$. Atoms in the sublevel $|i\rangle$ cannot decay to the $5P_{1/2}$ state. However, in the neighborhood of the anticrossings at 3800 G, atoms in sublevel $|i\rangle$ can be transferred to the sublevel $|j\rangle$ by the electric quadrupole coupling between the electrons and the nucleus. Since the atoms in the sublevel $|j\rangle$ can decay to the $P_{1/2}$ state, an increase in the 7947-Å fluorescence may be produced by the anticrossing. We make no attempt to analyze the polarization of the 7947-Å fluorescence. It is worth pointing out that conventional level crossings at nonzero magnetic fields in the 4^2D state cannot be observed since the coherence between the crossing sublevels is lost in the intervening stages of cascading.[5] However, cascade anticrossings, which result from population unbalance between magnetic sublevels can be readily observed. Further details about cascade anticrossing spectroscopy and its relationship to other anticrossing experiments[6] will be published later. Some typical anticrossing signals are shown in Fig. 2.

The magnetic fields $H(m_F)$ at the centers of the anticrossing resonances can be calculated from the Hamiltonian (6) and they are

$$H(m_F) = \frac{2\mu_I}{I}\left\{-\tfrac{2}{3}(m_F-3)\langle r^{-3}\rangle_l - \tfrac{1}{6}(m_F-3)\langle r^{-3}\rangle_d + \tfrac{5}{21}(m_F-\tfrac{3}{2})\langle r^{-3}\rangle_c\right\}$$
$$-\frac{Qe^2}{112}\langle r^{-3}\rangle_q \frac{h}{\mu_B}\frac{[36m_F^2 - 116m_F + 129 - 12I(I+1)]}{I(2I-1)} + H_0\left(1 - \frac{5\mu_I}{6I\mu_B}\right) \qquad (10)$$

for the anticrossings in the neighborhood of 3800 G. Here H_0 is the crossing field in the absence of hyperfine structure. The cascade anticrossing signals may be shown to be of Lorentzian form, with a full width at half-maximum (in gauss) of

$$\Delta(m_F)=\left[\left(\frac{h}{2\pi\tau}\right)^2+|2\langle i|\mathcal{H}_q|j\rangle|^2\right]^{1/2}\frac{5}{6\mu_B}, \qquad (11)$$

where τ is the radiative lifetime of the $4D$ state. Expressions similar to (10) and (11) can be written down for the crossings near 4800 G.

We have measured the positions of several anticrossing signals in Rb^{85} and Rb^{87}, as shown in Table I. In combination with the low-field measurements of $A_{3/2}$ and $A_{5/2}$, we have eleven independent measurements. We have made a least-squares fit of these measured quantities with five unknown parameters, namely, the fine structure constant ζ and $\langle r^{-3}\rangle_i$ where $i=l$, d, q, and c. The best fitted values of the fine-structure splitting $[(L+\frac{1}{2})\zeta]$ and $\langle r^{-3}\rangle_i$ are given in the abstract. The values of the magnetic dipole coupling constants $A_{3/2}$ and $A_{5/2}$ and those of the anticrossing fields calculated from these values of ζ and $\langle r^{-3}\rangle_i$ are shown in the fourth column of Table I to show the consistency of our least-squares fit.

Lindgren[7] has recently calculated the values of $\langle r^{-3}\rangle_i$ for higher D states ($6D$ and $7D$) of Rb. It is interesting to note that the preliminary results of his first-order calculations show that $\langle r^{-3}\rangle_d$ is negative, and $\langle r^{-3}\rangle_c$ has large negative values for both of these states. Better measurements of the widths of the anticrossing signals should yield a more precise value of $\langle r^{-3}\rangle_q$ for comparison with Sternheimer's recent calculation.[8]

Cascade anticrossing spectroscopy can be readily extended to many other states of alkali and other atoms to measure fine structures and to determine the complete hyperfine Hamiltonians.

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Specific Heat of Solid ^{3}He below 25 mK

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The specific heat of solid ^{3}He has been measured between 1 and 25 mK. The results indicate that the ordering of the nuclear spins takes place at a lower temperature than was anticipated theoretically, and with an unexpected temperature dependence.

In the millikelvin temperature range, phonons make a negligible direct contribution to the thermodynamic properties of solid ^{3}He. The spin degrees of freedom of the nuclei are, therefore, all that remain. The ^{3}He nuclei have spin $\frac{1}{2}$ and are arranged in a bcc lattice. Thus it is an exceptionally simple system. Nonetheless, a description of it in terms of the Heisenberg Hamiltonian has been found to be inadequate.[1] Inclusion of triple exchanges in addition to the pair exchanges of the Heisenberg model has improved the agreement with experiment.[2] No experiments up to this time have included measurements through the critical temperature, so that no direct evidence concerning the transition to the ordered state has been available. We report here on measurements of the specific heat C_v through the anticipated ordering temperature. They indicate that the transition is substantially different from that in other "simple" magnetic systems. They cannot be reconciled with earlier measurements of pressure in high magnetic fields within the framework of the triple-exchange theory.

The measurements were performed, for the