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Determination of Hyperfine Structures in Ground and Excited Atomic Levels by Level-Crossing Optogalvanic Spectroscopy: Application to ^{89}Y

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This new technique in optogalvanic spectroscopy yields Doppler-free resonances when degeneracies in atomic energy levels are removed by a magnetic field, and also when the interval between levels matches the mode spacing of the laser. g_J factors and magnetic hyperfine interaction constants were determined for lower and upper levels of the transition at 619.2 nm in YI. The value $A = (+)89.6(9)$ MHz for the upper level $4d5s(a\,^3D)5p\,^2D_{3/2}{}^0$ is new; the remaining determinations agree with published values.

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The change in impedance of a gas discharge caused by irradiation of the plasma with laser light tuned to some atomic transition (the optogalvanic effect) has become an important and sensitive technique for detecting atomic transitions in laser spectroscopy.[1] It has recently been demonstrated that a change in the optogalvanic signal accompanies the removal, by a small magnetic field, of the *zero-field degeneracy* of Zeeman levels.[2] We report here the location of *level crossings at finite fields* by the observation of changes in the optogalvanic signal as a function of applied magnetic field. The direction of the magnetic field must be crossed with (or oblique to) the direction of polarization of the laser light. We apply the phenomenon as a new Doppler-free technique to the determination of hyperfine structures in both ground and excited atomic levels. The experiments were carried out on the $4d5s^2$-$^2D_{3/2}$–$4d5s(a\,^3D)5p\,^2D_{3/2}{}^0$ 619.2-nm transition in atomic yttrium.

The laser is tuned to some frequency within the Doppler absorption profile of the transition. Both upper and lower states have hyperfine structures much smaller than the Doppler width: Both structures undergo decoupling in the magnetic field, and crossings between hyperfine levels occur for the upper and for the lower states. At values of the magnetic field where crossings occur between levels differing by ± 2 in m quantum number, the

laser light coherently couples one lower level with a pair of upper levels or one upper level with a pair of lower levels. The coherence is lost as the degeneracy is removed and the total population of the upper level changes provided the interaction is nonlinear.[3,4] The change of total population leads to a change in optogalvanic signal.[2,5,6] Two types of interaction have been distinguished as contributing to the change of population: A "population effect" whose width depends on the spectral width of the laser if that is larger than the homogeneous width of the atomic transitions (as it is in our experiments) and a "Zeeman coherence effect," whose width represents the combined homogeneous widths of the upper and lower levels. In our experiments we did not undertake an analysis of the line profile into these components, but we obtained evidence of their separate contributions.

The 619.2-nm yttrium line was chosen for initial investigation because it is a relatively strong transition based on the ground state and lying within the tuning range of rhodamine 6 G, and because the Zeeman energy-level structure is particularly convenient: Over the range of magnetic fields 0–120 G, two crossings having $|\Delta m| = 2$ occur in the ground levels and two in the excited levels. Furthermore, the optogalvanic signals were found to show a strong zero-field level-crossing resonance. The spectroscopic data for the system are summarized in Table I.

The yttrium atoms were sputtered from a cathode of naturally occurring yttrium (100% ^{89}Y) into a discharge maintained in argon at a pressure of about 1 Torr. The discharge current was about 5 mA and the optogalvanic signals typically 10 μA in the sense of enhancing the discharge current. Stray magnetic fields were reduced to less than 20 mG. The laser was a cw tunable laser running multimode at a power density of about 30 mW mm^{-2} distributed over all modes. The width of the individual modes (5–7 MHz) was greater than the pressure-broadened width of the upper level ($\approx$ 2 MHz) but much less than the Doppler width ($\approx$ 600 MHz). The separation of longitudinal modes was determined to be 401 $\pm$ 2 MHz.[13] The resonances were detected by modulating the magnetic field in addition to chopping the laser beam, and by tuning the phase-sensitive detector to the difference of the two modulation frequencies.

Six resonances, of which one is illustrated in Fig. 1, were found between 0 and 160 G, and a question of identification arises. We shall show later that the four resonances below 150 G correspond to the positions of the level crossings and serve to determine the values of $|A/g_J|$ (A is the magnetic dipole hyperfine interaction constant). The two resonances above 150 G, which were weaker than the other four, derive from interaction of the atoms with more than one longitudinal mode of the laser and are similar to the multimode saturation resonances found in the optogalvanic signals from transitions in Zr.[13] Taken together, the six resonances (i) provide internal checks on the interpretation and (ii) determine separately the A and g_J factors of the upper and lower states.

We draw attention to the steepness of the cen-

TABLE I. Spectroscopic data for the 619.2-nm transition in ^{89}YI ($I = \frac{1}{2}$).

	Term	Wave number (cm^{-1})	Radiative lifetime (ns)	g_J	A (MHz)
Ground state (a)	$4d5s^2\,^2D_{3/2}$	0	•••	0.798(2)[b]	−57.217(15)[e]
				0.799 27(11)[c]	(−)57.2(5)[d]
				0.800(8)[d]	
Excited state (z)	$4d5s(a\,^3D)5p\,^2D_{3/2}{}^0$	16 146	175(10)[a]	0.797(3)[b]	$\approx$ (+)100[f]
				0.801(8)[d]	(+)89.6(9)[d]

[a] Ref. 7.
[b] Ref. 8.
[c] Ref. 9.
[d] This work.
[e] Ref. 10.
[f] Estimated (Ref. 11) from measured hyperfine splitting in $z\,^2D_{5/2}{}^0$ state assuming $A(z\,^2D_{3/2}{}^0)/A(z\,^2D_{5/2}{}^0)$ to be the same as that of the corresponding states in scandium (Ref. 12).

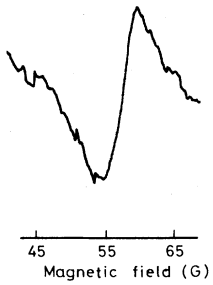

FIG. 1. Field-dependent optogalvanic signal showing the level-crossing B_1^a in the lower ($a\,^2D_{3/2}$) state of the transition 619.2 nm in ^{89}Y. Magnetic field modulation. The derivative signal appears on a sloping background, representing the direct effect of the magnetic field on the discharge current. The amplitude of the level-crossing signal represents a change of about 1.5% in the optogalvanic current.

tral portion of the experimental curve in Fig. 1, which, considered in relation to the outer portions, echoes the characteristically pointed shape of the zero-field resonances in Zr.[2] This feature is consistent with the superposition of a narrow Zeeman-coherence resonance on a broader population-effect resonance. The full width at half height is about 7 G (8 MHz), which is comparable with the width of individual modes of the laser, but very much less than the Doppler width.

We identify the first four resonances in Table II as two related pairs of level-crossing resonances because the ratio of the fields at which these resonances occur is 0.748 for the first pair and 0.743 for the second, and these values are satisfactorily close to the value $5^{1/2}/3 = 0.745$ predicted by the Breit-Rabi formula for $|\Delta m| = 2$ level crossings in systems having $J = \frac{3}{2}, I = \frac{1}{2}$. The values of $|A/g_J|$ deduced from the positions of the first pair of resonances are essentially the same as the accurately known value of $|A/g_J|$ for the $a\,^2D_{3/2}$ ground state[9,10] (Table II), and we identify this pair, B_1^a and B_2^a, as level crossings arising from the ground state. The second pair of resonances, B_1^z and B_2^z, we attribute to level crossings in the $z\,^2D_{3/2}{}^0$ excited state. This interpretation was confirmed by the results of *fluorescence studies* under the same conditions: Each member of the second pair of resonances could be analyzed into two superimposed components, one of which inverted with change through $\pi/2$ of the polarizer. This behavior is characteristic of the fluorescence from excited atoms *aligned* by irradiation with linearly polarized light. The component which did not invert under rotation of the polarizer is attributable to the effect of irradiation on the *combined population* of the crossing levels.

We identify the pair of high-field resonances (near 150 G) as multimode saturation resonances because the fields at which they occur correspond to Zeeman splittings which match very closely the mode separation of the laser (401 ± 2 MHz). (This analysis was performed by using our determined values of $|A/g_J|$ and g_J factors obtained from other sources[8,9].) The first member of this pair, B_3^z, arises from the splitting of the $F = 1$, $m = \pm 1$ levels in the excited state, and the second, B_3^a, from the splitting of the corresponding levels in the ground state. The identification of these resonances as multimode saturation resonances is further supported by the observation that they vanish when the laser is stabilized so as to lengthen appreciably the time between mode hops. Addi-

TABLE II. Observed positions of level crossing (LC) and multimode (MM) saturation resonances in the 619.2-nm optogalvanic signal from ^{89}Y.

Position (G)	Ratio	$\lvert A/g_J\rvert$ (MHz) This work	$\lvert A/g_J\rvert$ (MHz) ABMR[a]	$\Delta\nu$[b] (MHz)	Identification[c]
57.2(5)	0.748(8)	71.6(6)	71.59(2)		LC B_1^a
76.5(5)		71.4(4)	71.59(2)		LC B_2^a
89.2(5)	0.743(7)	111.7(6)	...		LC B_1^z
120(1)		112.0(9)	...		LC B_2^z
150.5(1.0)				399(3)	MM B_3^z
157(1)				401(5)	MM B_3^a

[a]Atomic-beam magnetic resonance studies; Refs. 9 and 10.

[b]Frequency of Zeeman splitting at position of observed resonance; determined with use of $|A/g_J|$ values from column 3 and g_J factors from Refs. 8 and 9.

[c]Superscript a refers to ground state, and z to excited state.

tional multimode resonances corresponding to $|\Delta m_J| = 2$ within the $F = 2$ complexes would have been expected at higher fields, but this region was not explored. Resonances $|\Delta m_J| = 2$ between $F = 1$ and $F = 2$ were expected near 145 G (lower level) and 119 G (upper level). There was, indeed, evidence of a weak resonance near 145 G, but this was not investigated in detail. The resonance near 119 G would have been obscured by the level crossing B_2^z.

From the measured positions of the six identified resonances (Table II) we determine the A factors of the excited and ground levels directly by setting the Breit-Rabi expression for the splitting of the Zeeman $F = 1, m = \pm 1$ levels at B_3 equal to the mode separation of the laser, i.e., $A[4x_3 - (1 - x_3 + x_3^2)^{1/2} + (1 + x_3 + x_3^2)^{1/2}] = 401$ MHz, where $x_3 = 5^{1/2}B_3/4B_1$ or $3B_3/4B_2$. We note that the A factors determined in this way depend only on the *ratio* of pairs of magnetic field values and on the frequency difference between adjacent modes of the laser. In particular, they do not rely on prior knowledge of g_J, and the accuracy is determined by the accuracy with which ratios of magnetic field values have been measured, not absolute values.

Mean values of the A factors, together with the g_J factors which the analysis also yields, are presented in Table I. The sign of the A factor for the $a\,^2D_{3/2}$ ground state is known to be negative from atomic-beam magnetic resonance studies.[10] As for the $z\,^2D_{3/2}{}^0$ excited state, we note that in scandium, for which the term structure closely resembles that of yttrium, the A factor of the corresponding $3d4s(a\,^3D)4p\,^2D_{3/2}{}^0$ level has opposite sign to the $3d4s^2\,^2D_{3/2}$ ground level; also note that the fine-structure splitting of the yttrium $z\,^2D_{3/2,\,5/2}{}^0$ states is inverted, whereas that for the $a\,^2D_{3/2,\,5/2}$ states is normal. The A factor for the yttrium $z\,^2D_{3/2}{}^0$ level is therefore very likely to be positive.

Our A factor for the $a\,^2D_{3/2}$ ground state is in good agreement with the very accurate atomic-beam magnetic resonance result of Fricke, Kopfermann, and Penselin[10] (Table I). The value for the $z\,^2D_{3/2}{}^0$ state appears to be the first reported measurement for that level. It is fairly close to a value estimated by Kuhn and Woodgate[11] from their measured A factor for the $z\,^2D_{5/2}{}^0$ state, assuming a value of $A(z\,^2D_{3/2}{}^0)/A(z\,^2D_{5/2}{}^0)$ found for the corresponding states in scandium.[12]

We conclude that level-crossing optogalvanic spectroscopy offers a new high-resolution spectroscopic technique for determining hyperfine structures in both excited and ground atomic levels. When the exciting laser is run multimode and the frequency separation of the modes is known accurately, the technique can yield hyperfine interaction constants independently of the Landé g_J factors, and only *relative* values of the magnetic fields need to be determined accurately.

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