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LEVEL-CROSSING OPTOGALVANIC SPECTROSCOPY

P. Hannaford, D.S. Gough and G.W. Series*

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Résumé - La spectroscopie optogalvanique est utilisée pour la détection de croisements de niveaux en champ magnétique nul ou de valeur finie. Cette méthode offre une technique sensible, sans effet Doppler, pour déterminer la structure hyperfine des niveaux profonds et élevés impliqués dans les transitions atomiques.

Abstract - Optogalvanic spectroscopy is applied to the detection of level-crossings at zero and finite magnetic fields. The method provides a sensitive, Doppler-free technique for determining hyperfine structures in both lower and upper levels of atomic transitions.

I - INTRODUCTION

The detection of changes in the polarization properties of fluorescent light when two sublevels become degenerate, either at zero or finite magnetic field, has become a well-established technique for determining spectroscopic quantities such as hyperfine structure factors, Stark shifts, radiative lifetimes and collisional disalignment cross sections of excited atomic levels /1/. The resolution of the technique is determined by the homogeneous width of the crossing sublevels (typically 1-10 MHz) and thus allows very small splittings to be determined with a high degree of precision.

In this paper we describe the application of optogalvanic spectroscopy to the detection of level-crossings. The mechanism for detecting the crossings differs from that normally employed in fluorescence experiments. The optogalvanic current in a gas discharge at pressures of the order of 1 torr tends to behave as an isotropic probe of atomic levels and is normally sensitive only to the populations of the levels connected by the laser /2/, in contrast with fluorescent light which is sensitive to alignment and orientation of the levels as well as population. (Atomic alignment might be expected to be detectable using optogalvanic spectroscopy under certain discharge conditions /3,4/, and indeed there has been a very recent report /5/ of detection of alignment in optogalvanic signals from a neon discharge operated in a magnetic field of about 1 kG /)). In optogalvanic experiments where the light source is a cw laser, one may exploit the fact that the transition may be saturated or partially saturated. Under these conditions, there may occur in the vicinity of a level-crossing a redistribution of population between the upper and lower levels, which is reflected as a resonance /6,7/ in the optogalvanic current. Level-crossing saturation resonances of this type had previously been observed in fluorescence/8,9/ and absorption /10/ experiments, and it was predicted /11/ that they should also be observed with optogalvanic detection. A theoretical treatment of the level-crossing optogalvanic resonances, based on a simple rate-equation approach, has been reported recently by Dodd /12/.

The redistribution of population that occurs in the vicinity of a level-crossing under conditions of saturation can be easily understood by considering the simple case of a zero-field crossing for a $J_a=0$ to $J_b=1$ transition (Fig.1). The laser light is assumed to be linearly polarized in a direction perpendicular to the magnetic field direction so that the $\sigma^+$ and $\sigma^-$ transitions are stimulated. For simplicity it is assumed that these transitions are completely saturated by the...
Fig. 1 - Redistribution of atomic population that can occur in the vicinity of a zero-field level-crossing under conditions of saturation. The three regions correspond to (a) high magnetic field, where two different velocity groups ($2n$ atoms) interact with the laser, (b) low field, where only one group ($n$ atoms) interacts, and (c) zero field, where there is a coherent superposition of $\sigma^+$ and $\sigma^-$ transitions.

At high magnetic fields (Fig. 1a) the splitting of the $m_b = \pm 1$ sublevels is such that the $\sigma^+$ and $\sigma^-$ transitions are resolved with respect to their homogeneous width $2\Gamma_{ab}$ (where $\Gamma_{ab}$ is the relaxation rate of the optical coherence, in the notation of Decamps, Dumont and Ducloy/8/), and two distinct "holes" are burned into the velocity distribution of the atoms by the laser. Under conditions of saturation, the laser field equalizes the populations in the upper and lower levels for each of the $\sigma^+$ and $\sigma^-$ transitions. Thus if there are $n$ atoms interacting with the laser within each velocity group, the total number of atoms in the $J_b = 1$ upper level at equilibrium will be $n/2 + n/2 = n$. At low magnetic fields (Fig.1b) where the $\sigma^+$, $\sigma^-$ splitting is smaller than $2\Gamma_{ab}$, atoms in the same velocity group are stimulated to make $\sigma^+$ and $\sigma^-$ transitions, and only a single hole is burned into the velocity distribution; so that if the splitting is greater than the homogeneous width of the upper level $\Gamma_b$, the laser field equalizes the populations in each of the three sublevels $m_b = 0$, $m_b = +1$ and $m_b = -1$. The total number of atoms in the upper level is then $n/3 + n/3 = 2n/3$. At zero field, the $m_b = \pm 1$ sublevels are degenerate (Fig. 1c) and there is a coherent superposition of the $\sigma^+$ and $\sigma^-$ transitions which is equivalent to a single $\sigma$ transition; so that $n/2$ are excited to the upper level. The same result is found by assuming linear-$\sigma$ polarization and including the resulting upper-level Zeeman coherence in the calculation. Thus as the magnetic field is swept through a level-crossing at zero field, the population in the upper level (and hence the optogalvanic current) exhibits a resonance characterized by the homogeneous width of the transition $2\Gamma_{ab}$ (or $2\rho_{ab}/g_{ppB}$ in units of magnetic field), upon which is superimposed a narrower resonance characterized by the homogeneous width of the upper level $\Gamma_b$. For the pressure conditions in a rare-gas discharge, the width $2\Gamma_{ab}$ is expected to be much greater than $\Gamma_b$ because the optical coherence is more sensitive to collisions than is the Zeeman coherence $g/9$. The broad resonance is sometimes called the population effect or hole-burning effect and the narrow resonance the Zeeman coherence or non-linear Hanle effect. The strength of the population effect resonance (ratio of size of dip to high-field signal) predicted by this simple model is $S = (n - 2n/3)/n = 0.33$ and the strength of the Zeeman coherence resonance $S = (2n/3 - n)/2n/3 = 0.25$.

In practice the strengths of the resonances are reduced by collisional relaxation between the $m_b = 0$, $\pm 1$ sublevels/12/ and also by the fact that there is incomplete saturation in the wings of the frequency interaction profile and in the wings of the spatial profile of the laser beam. For transitions with certain combinations of $J$-values, one might expect to see additional effects, such as resonances due to
crossings in the lower level, effects of optical pumping into sublevels which do not interact with the laser, and very high-order effects.

A schematic diagram of the experimental arrangement used for optogalvanic detection of level-crossings is shown in Figure 2. An atomic vapour of the element of interest (e.g. Zr) is produced by cathodic sputtering in a low-pressure neon or argon discharge and excited in the presence of a magnetic field by a single-mode cw dye laser. The laser light is sufficiently strong to saturate, or partially saturate, the atomic transition and is linearly polarized in a direction perpendicular to the magnetic field.

The lock-in can be referenced either to (a) the modulation frequency of the laser \( f_2 \), which allows the total optogalvanic current to be detected as a function of applied field \( B \); or to (b) the sum frequency \( f_1 + f_2 \), where \( f_1 \) is the frequency of a small modulated component \( B_0 \sin 2\pi f_1 t \) applied to the field coils. The double-modulation mode of detection allows a derivative signal of the field-dependent part only of the optogalvanic current to be detected as a function of \( B \).

II - ZERO-FIELD LEVEL CROSSINGS.

Figure 3 shows a zero-field level-crossing resonance recorded in the optogalvanic current for the 613.5 nm \( (a^3F_2 - z^3F_2^0) \) transition in Zr at three laser power densities. The resonance vanishes when the direction of polarization of the laser light is rotated through \( \pi/2 \) so that it coincides with the magnetic field direction, and it remains unchanged, both in shape and strength, irrespective of the direction of the discharge current relative to the direction of polarization. The strength of the resonance \( S \) (ratio of size of dip to high-field signal) is strongly dependent on the power density of the laser (Fig. 4), exhibiting typical saturation behaviour. For the particular conditions in Fig. 3 (0.8 torr neon, single-mode laser with beam diameter \( \approx 1 \) mm), \( S \) has a saturation value of \( S(\infty) = 0.14 \) and the laser power required to reach \( S(\infty)/2 \) is \( P(\infty) = 0.3 \) mW. The actual values of \( S(\infty) \) and \( P(\infty) \) are quite sensitive to gas pressure: over the range 0.5 - 3 torr Ne, \( S(\infty) \) varies from 0.17 to 0.06 and \( P(\infty) \) from 0.15 to 0.45 mW. The width of the resonance at very low laser power approaches a value of 4 G, or 3.8 MHz in frequency units (using the Landé g-factor for either the upper or lower level). This value is greater than the pressure-broadened homogeneous width of the upper level, \( \gamma_0/2\pi = 1.2 \) MHz, as determined from the linear Hanle effect in fluorescence, but very much smaller than the Doppler width of the transition (about 600 MHz) and the homogeneous width of the transition expected at the pressures used in the discharge. It is also greater than the short-term (< 10 ms) bandwidth of the laser, < 2 MHz, and remained unchanged when a frequency-stabilized laser having a long-term bandwidth of
0.5 MHz was employed. The shape of the resonance is fairly close to Lorentzian at low laser power (Fig. 3(a)), but becomes very non-Lorentzian as the power is raised: the resonance broadens appreciably in the wings and the central region sharpens (Fig. 3(c)). An interpretation of this behaviour is that at low laser power the resonance consists predominantly of Zeeman coherence components from the upper and lower levels, while at higher powers an additional narrow Zeeman coherence component is coupled to the population of the levels by the laser field. The additional component could be a $\Delta m = 4$ coherence which evolves at four times the Larmor frequency and so is destroyed by a weaker magnetic field. A detailed theoretical analysis for the case of a $J_a = 2$ to $J_b = 2$ transition is currently being performed. The reason for the difference between the width of the optogalvanic resonance at low laser power (3.8 MHz) and the pressure-broadened homogeneous width of the upper level ($\approx 1.2$ MHz) is not clear and will be the subject of further study.

Zero-field optogalvanic resonances have also been studied for a large number of other Zr transitions lying within the tuning range of rhodamine 6G. These transitions originate from either the ground or metastable states and provide a wide range of transition probabilities and combinations of $J$-values and $g$-factors. Quite large values of $S (>0.05)$ are found in all cases, provided the power density of the laser is sufficiently high to saturate the transition. The saturation curves for the 614.3 nm ($a^3F_3 - z^3F_2^0$) and 612.7 nm ($a^3F_4 - z^3F_2^0$) transitions are very similar to Fig. 4, yielding $S(\bar{m})$ and $P(S)$ values essentially the same as for the 613.5 nm ($a^3F_2 - z^3F_2^0$) transition. The saturation curve for the 602.5 nm
The intercombination line also yields a similar value of $S(m)$, but $p(S)$ is higher by a factor of 25 to 30. The high $p(S)$-value can be explained in terms of the very small branching ratio (0.03) for the 602.5 nm transition: about 30 times more laser power is required to achieve the same level of saturation. Similarly, large values of $p(S)$ are found for other Zr transitions having small branching ratios, while for transitions for which $J_a \neq J_b$, the $S(m)$-values are smaller by factors of 1.5 to 2.5. Strong zero-field optogalvanic resonances have also been observed for transitions in yttrium and samarium, and recently by Barbieri et al. for transitions in neon and calcium.

III - LEVEL-CROSSINGS AT FINITE FIELDS.

In some systems it is possible to have crossings between $|\Delta m|=2$ sublevels which originate from different neighbouring levels, and in particular from different hyperfine levels. Levels of yttrium provide a good example of this (Fig. 5). Yttrium consists of 100% $^{89}\text{Y}$ ($I = \frac{4}{2}$), which has a small magnetic moment (-0.1368 n.m.), and there are a number of suitable transitions that lie within the tuning range of rhodamine 6G. The particular transitions of interest are those between the ground levels $4d5s^2 \zeta^2D_0^2$ and levels within the terms $4d5s5p \zeta^2D_0^2$ and $\zeta^4P_0^0$. The hyperfine structures for these transitions are normally unresolved in the presence of Doppler broadening /14/, and the only accurate hyperfine structure data available prior to this investigation have been the atomic-beam magnetic resonance measurements on the $a^2D_3/2$ ground levels /15/.

The first system investigated was the $619.2$ nm ($a^2D_3/2 - z^2D_3/2$) transition, for which, since $J = 3/2$ and $I = \frac{4}{2}$, two $|\Delta m|=2$ crossings are to be expected in both the upper and lower levels (Fig. 5). Figure 6 shows the level-crossing resonances observed in the range 0-150G (using the double-modulation mode of detection - Fig. 2). A strong zero-field resonance is found, followed by four weaker resonances, labelled $B_1^a$, $B_2^a$, $B_1^z$, and $B_2^z$. The ratio of the magnetic field values at which the first two weaker resonances occur, $B_1^a/B_2^a$, and the corresponding ratio $B_1^z/B_2^z$ for the second pair of resonances, are both very close to the value $5^2/3 = 0.7454$ predicted by the Breit-Rabi formula for systems having $J = 3/2$, $I = \frac{4}{2}$.

![Fig. 5 - Zeeman energy-level diagram for the upper and lower levels of the 619.2 nm ($a^2D_3/2 - z^2D_3/2$) transition in $^{89}\text{Y}$ $I (I = \frac{4}{2})$. Lower-level parameters: $A = -57.23$ MHz, $g_j = 0.79927$. Upper-level parameters: $A = 89.9$ MHz, $g_j = 0.797$. The $|\Delta m|=2$ level-crossings are indicated by open circles. The vertical lines represent the paths by which the $|\Delta m|=2$ crossings can be reached.]
The first two resonances appear at the same magnetic field values for other Y I transitions (602.3, 622.3 and 608.3 nm) having the same lower level, and these are therefore identified as arising from crossings in the \( \alpha^2D_{3/2} \) level. The second pair is interpreted as arising from crossings in the upper level, \( \alpha^2D_{5/2} \).

Level-crossings at finite magnetic fields have also been studied for the 622.3 nm (\( \alpha^2D_{3/2} - \alpha^2D_{5/2} \)), 608.3 nm (\( \alpha^2D_{3/2} - \alpha^2D_{5/2} \)), 602.3 nm (\( \alpha^2D_{3/2} - \alpha^2D_{5/2} \)) and 613.3 nm (\( \alpha^2D_{3/2} - \alpha^2D_{5/2} \)) transitions of Y I. The \( J = 5/2 \) levels have four \( |\Delta m| = 2 \) crossings, while the \( J = 7/2 \) level does not have a crossing. The resonances observed for the 608.3 nm transition are therefore due solely to level-crossings in the \( \alpha^2D_{3/2} \) ground level, and so this transition was useful for identification of \( \alpha^2D_{3/2} \) resonances in other transitions.

The location of the level-crossings allows \( |A/I| \) to be determined for each level, where \( A \) is the magnetic hyperfine interaction constant. The \( g_\| \)-factors are usually known from earlier work, although not always with the desired precision. They can, however, be determined independently, if required, using the optogalvanic method described in the following section. Furthermore the signs of the \( A \)-factors can be determined in some cases by studying the respective widths of the zero-field optogalvanic resonance with the laser tuned first to one side of the Doppler absorption profile, and then to the other. For the Y I 619.2 nm transition, for example, the zero-field resonance (Fig. 7) is broader by a factor of about 1.6 when the laser is tuned to the high frequency side of the Doppler profile (trace labelled b) than when it is tuned to the low frequency side (trace a). The major

The inset shows the hyperfine structure components at zero field for the 619.2 nm transition.
Table 1. Hyperfine interaction constants (A) in levels of Y I.

<table>
<thead>
<tr>
<th>Level</th>
<th>( g_J )</th>
<th>A (MHz)</th>
<th>This work (^a)</th>
<th>Other work (^d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 4d^5 )</td>
<td>( 2D_3/2 )</td>
<td>0.7993(1)(^b)</td>
<td>-57.23(3)</td>
<td>-57.217(15)</td>
</tr>
<tr>
<td>( 4d^5 )</td>
<td>( 2D_5/2 )</td>
<td>1.2003(2)(^b)</td>
<td>(-)28.77(5)</td>
<td>-28.749(30)</td>
</tr>
<tr>
<td>( 4d^5(3D) )</td>
<td>( 2D_3/2 )</td>
<td>0.797(3)(^c)</td>
<td>+89.9(2)</td>
<td></td>
</tr>
<tr>
<td>( 4d^5(3D) )</td>
<td>( 2D_5/2 )</td>
<td>1.203(5)(^c)</td>
<td>-218.7(3)</td>
<td>-190(12)(^e)</td>
</tr>
<tr>
<td>( 4d^5(3D) )</td>
<td>( 4D_3/2 )</td>
<td>1.22(3)(^c)</td>
<td>+39.5(1)</td>
<td></td>
</tr>
<tr>
<td>( 4d^5(3D) )</td>
<td>( 4D_5/2 )</td>
<td>1.38(3)(^c)</td>
<td>-133.7(1)</td>
<td></td>
</tr>
</tbody>
</table>

\(^a\) Indicated uncertainties do not include contribution from \( g_J \) (col. 3).
\(^b\) Ref. 16. \(^c\) Ref. 17. \(^d\) Ref. 15. \(^e\) Ref. 14.

Hyperfine components in the 619.2 nm transition are the \( F = 1 \) to \( F' = 1 \) and \( F = 2 \) to \( F' = 2 \) (see inset to Fig. 7). The \( g_F \)-factors for the hyperfine levels \( F = 1 \) and \( F' = 1 \) are both close to 1.0, while those for the levels \( F = 2 \) and \( F' = 2 \) are both close to 0.6. If, on the one hand, the \( A \) - factors for the levels \( 2D_3/2 \) and \( 2D_5/2 \) (\( A^a \) and \( A^a \)) have different signs, then the splitting of the \( 1 \to 1 \) and \( 2 \to 2 \) hyperfine components is \( |2A^a| + |2A^a| = 300 \) MHz, which is comparable with the Doppler width (= 600 MHz), and one would expect the width of the zero-field resonance to be governed essentially either by \( g_F = 2 = 0.6 \) or by \( g_F = 1 = 1.0 \), depending on whether the laser is tuned close to the \( 2 \to 2 \) component or the \( 1 \to 1 \) component. On the other hand, if \( A^a \) and \( A^a \) have the same sign, then the splitting of the \( 1 \to 1 \) and \( 2 \to 2 \) components is only \( |2A^a| - |2A^a| = 70 \) MHz and one would expect the width of the zero-field resonance to be relatively insensitive to laser tuning. The observed ratio of widths (1.6) is close to the ratio \( g_F = 1/g_F = 2 = 5/3 \) and is compatible only with the assignment \( A^a < 0, A^a > 0 \).

The \( A \)-factors determined in this study are summarised in Table 1, together with the results of other workers. The \( g_J \)-factors used in the analysis are also shown. The uncertainties in the \( A \)-factors are typically 1 part in 500, if the contribution from \( g_J \) is not included.

IV - MULTIMODE SATURATION RESONANCES

When the laser source is run multimode, additional resonances are observed in the optogalvanic current at magnetic fields where the Zeeman splitting in either the upper (\( 2g_\text{ub}B/h \)) or lower (\( 2g_\text{lb}B/h \)) levels is equal to a multiple of the mode separation of the laser (Fig. 8). Under these conditions, for one and the same velocity group of atoms, the \( \sigma^+ \) transition is excited by one mode and the \( \sigma^- \) transition excited by another mode. When the \( \sigma^+ \) and \( \sigma^- \) transitions are saturated...
or partially saturated by the laser, their simultaneous excitation can lead to a reduction in the number of atoms in the upper level, and hence to a resonance in the optogalvanic current. The location of these multimode saturation resonances allows Landé g-factors to be determined for the upper and lower levels, provided the mode separation of the laser is known. Similar multimode saturation resonances have previously been observed in fluorescence for transitions in neon /18,19/.

In the publication in which we first reported these multi-mode resonances /20/ we attributed their occurrence to the population effect, not to the Zeeman coherence effect, on the grounds that the coherence time of our laser was short compared with the lifetimes of the atoms studied. As a result of further experiments it now appears likely that, under the conditions in which the multimode resonances appear, the short-term coherence time of the laser (which is the significant quantity) is, in fact, considerably longer than the lifetime of the excited atoms, and our present view is that the Zeeman coherence effect, not the population effect, is the dominating mechanism. This was the interpretation offered by the authors of the work in neon /18,19/. In those experiments also the coherence time of the laser was long compared with the lifetimes of the atomic levels.

The trace shown in Fig. 8 was recorded in the optogalvanic current for the 612.7 nm \( \left( ^{3}P_{4} - ^{3}F_{2} \right) \) transition in Zirconium, using a multimode ring laser as the source. The first resonance is the familiar zero-field signal, and the other two (at 66 and 132 G), which vanish when the laser is running single-mode, are the nearest-mode and next-nearest-mode saturation resonances, respectively. The g-factors for the upper and lower levels of the 612.7 nm transition are almost identical \((g_a = 1.25 /20/ \text{ and } g_b = 1.24987 /21/)\) and consequently each of the observed multimode resonances for this transition represents an unresolved superposition of resonances associated with the upper and lower levels. The upper- and lower-level resonances were found to be resolved for certain \( \text{Zr I} \) transitions such as the 613.5 nm \((g_a = 0.66981, g_b = 0.696)\) and 614.0 nm \((g_a = 1.26472, g_b = 1.51)\).

The multimode saturation technique has recently been applied, using a linear dye laser having accurately determined mode separation, to obtain Landé g-factors for a number of levels in \( \text{Zr I} /20/ \) and \( \text{Y I} /7/\).

V - SUMMARY AND CONCLUDING REMARKS

The level-crossing optogalvanic technique described in this paper allows small hyperfine interaction constants to be determined, both in magnitude and sign, for the upper and lower levels of certain atomic transitions. The optogalvanic mode of detection is highly sensitive, allowing the study of very weak transitions, and it should also allow the study of highly-excited atomic states that are difficult or impossible to study by optical detection methods. The use of magnetic-field scanning in these experiments allows hyperfine splittings to be determined with a high level of precision (typically one part in 500 in the present investigation). The technique is well suited to the study of levels in refractory metal atoms, which can be readily produced by sputtering in a rare-gas discharge. Finally, the technique does not require a laser with frequency stabilization or frequency-scanning facilities.

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