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Observation of modulation in the transfer of orientation by collisions of the second kind

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Résumé. — On a observé les modulations lors du transfert collisionnel d'orientation entre atomes de mercure et de cadmium, sous l'application simultanée d'un champ magnétique constant et d'un champ tournant. On a étudié la dépendance de la profondeur de modulation en fonction du champ constant, et l'accord avec la théorie est satisfaisant.

Abstract. — Modulation has been observed in orientation transfer from mercury to cadmium under the simultaneous application of a static and a rotating magnetic field. The dependence of the depth of modulation on the static field has been investigated, and the agreement with theory is satisfactory.

1. Introduction. — The transfer of coherence from atoms of species A to atoms of a different species B has been extensively studied in Hg-Cd sensitized fluorescence:

\[
\text{Hg} (6 \, 1S_0) + h\nu (2 \, 537 \, \text{Å}) \rightarrow \\
\text{Hg} (6 \, 3P_1)
\]

\[
\text{Hg} (6 \, 3P_1) + \text{Cd} (5 \, 1S_0) \rightarrow \\
\text{Hg} (6 \, 1S_0) + \text{Cd} (5 \, 3P_1) + \Delta E (1.09 \, \text{eV})
\]

\[
\text{Cd} (5 \, 3P_1) \rightarrow \\
\text{Cd} (5 \, 1S_0) + h\nu (3 \, 261 \, \text{Å}).
\]

Previous work [1-3] has shown that quantum beats can be produced by a magnetic field \( H_1 \) rotating with angular frequency \( \omega_0 \) in a plane perpendicular to a steady field \( H \). In these experiments, the transfer of alignment, rather than orientation, was being studied.

Chéron [4, 5] showed that orientation is also transferred from the excited Hg to Cd atoms in the collision, giving rise to the Hanle effect. We wish to report experiments showing quantum beats in Hg-Cd sensitized fluorescence under the transfer of orientation.

2. Theoretical. — The intensity \( I_s \) of the sensitized fluorescence is calculated by expanding the density matrix of an excited atom in terms of irreducible tensor operators \( T_q^{(k)} \), where \( k = 0 \) (population), 1 (orientation) or 2 (alignment), and \(|q| \leq k\).

With \( H_1 \) applied, \( I_s(t) \) has been deduced [3] on the assumption that the collisions are isotropically distributed. The expression is rather complicated, but may be simplified by assuming (i) \( \omega_0 \) is much smaller than the decay constants \( \Gamma_A \) and \( \Gamma_B \) of the \( 3P_1 \) excited states of A and B, (ii) the range of \( |H| \) is restricted to be \( \leq \Gamma_B/\gamma \) (the range over which the Hanle effect in Cd occurs), where \( \gamma \) is the gyromagnetic ratio of the \( 3P_1 \) state. Since \( \Gamma_A \approx 20 \, \Gamma_B \), it follows that \( \gamma H \ll \Gamma_A \).

We find then approximately,

\[
I_s(t) \propto \sum_{k \leq q \leq k} \frac{\Gamma_A^{(k)} d^{(k)}_{q1} e^{(k)}_{q1} \exp i(q_1 - q_3) \omega_0 t a^{(k)}_{q1}(\beta) a^{(k)}_{q2}(\beta)}{\Gamma_B + i \nu_2 \gamma (H^2 + H_1^2)^{1/2}}.
\]

(1)

Here \( a^{(k)}_q \) and \( b^{(k)}_q \) are respectively the rate of excitation and the efficiency of detection of the component \( T_q^{(k)} \), \( \Gamma_A^{(k)} \) is the rate of transfer of component \( T_q^{(k)} \) from A to B, and \( \beta \approx \tan^{-1} H_1/H \).

3. Experiments. — The apparatus used is that shown in figure 2 of [3], to which reference is made for details. 2 537 Å radiation from a mercury lamp was circularly polarized, and incident on a mixture of Hg and Cd vapours. The sensitized fluorescence emitted perpendicular to the incident light passed through an analyser set to pass circularly polarized radiation. \(|\omega_0|/2 \pi \) was 20.5 Hz, which was negligible compared with \( \Gamma_A \) and \( \Gamma_B \), as required.
The results reported for sensitized fluorescence show the average of 15 runs. In between each, the polarizer and/or analyser was rotated through 90° to eliminate any effects due to residual ellipticity of polarization. A single run was also performed with the Hg lamp replaced by a Cd lamp (with the chlorine filter removed) to produce the corresponding signal in Cd resonance fluorescence. According to the theory, this should be the same shape as that for sensitized fluorescence.

4. Results. — To investigate modulation at \( \omega_0 \), \( H \) was directed parallel to the incident light. Eq. (1) then reduces to

\[
I(t) \propto 4 \Gamma_A^0(1 - 3A) - 6 \Gamma_A^0 \left\{ \pm \right\} \left( \pm \right) \left( \pm \right) \times \frac{b \Gamma_B}{\delta^2 + b^2 + \Gamma_B^2} \sin \omega_0 t + \frac{\delta b}{\delta^2 + b^2 + \Gamma_B^2} \cos \omega_0 t
\]

where \{ \pm \} = \pm 1 if the incident light is LH circularly polarized, and \{ \pm \} = \pm 1 if the analyser detects RH circularly polarized light. \( b = \gamma H \) and \( \delta = \gamma H - \omega_0 \approx \gamma H \). The functions \( A, D \) and \( E \) are those defined by Dodd and Series [6]. They are respectively

\[
b^2(4 \delta^2 + b^2 + \Gamma_B^2)/F, \quad b^2(2 \delta^2 - b^2 - \Gamma_B^2)/F
\]

and

\[
3 b^2 \Gamma_B \delta/F,
\]

where \( F = (\delta^2 + b^2 + \Gamma_B^2)(4 \delta^2 + 4 b^2 + \Gamma_B^2) \).

Modulation at \( \omega_0 \) arises from orientation transfer, and there is no contribution from the alignment. There are two signals in quadrature,

\[
b \Gamma_B/(\delta^2 + b^2 + \Gamma_B^2)
\]

which is symmetric in \( H \), and \( \delta b/(\delta^2 + b^2 + \Gamma_B^2) \) which is antisymmetric. These are the well-known Bloch functions. By careful setting of the phase control in the detection system, it was possible to isolate these two signals in resonance fluorescence and to check that they were of the expected form. With the same control settings, the symmetric and antisymmetric signals for sensitized fluorescence were observed (Fig. 1). These are to be compared with the coefficients of \( - \sin \omega_0 t \) and \( - \cos \omega_0 t \) in (2). Figure 2 shows the expected signal shapes. When the handedness of the incident polarization was reversed, the sign of the observed curves was reversed as expected.

To investigate modulation at \( 2 \omega_0 \), \( H \) was directed perpendicular to both the incident and the fluorescent beams, in which case

\[
I(t) \propto (\text{unmodulated terms}) +
\]

\[
+ \left\{ \frac{\delta}{\delta^2 + b^2 + \Gamma_B^2} + \Gamma_B^2 \right\} E \times \sin 2 \omega_0 t + \text{(modulation term at 4 \omega_0).}
\]

This time, there is no quadrature function in \( \cos 2 \omega_0 t \), and the one in \( \sin 2 \omega_0 t \) is a superposition of a symmetric function due to orientation transfer, and an antisymmetric one \( (E) \) due to alignment transfer.

Figure 3a shows the signal obtained, to be compared with the coefficient of \( - \sin 2 \omega_0 t \) in (3). An interesting feature is that the peak is slightly displaced from \( H = 0 \).

5. Discussion of results. — The mean lifetime of the Cd \( 5^3P_1 \) state is 2.25 \( \mu \)s, from which

\[
\Gamma_B = 444 \times 10^2 \text{ s}^{-1}
\]

Consequently, each increase of 1 in \( \delta/\Gamma_B \) corresponds to a change of about \( 3.4 \times 10^{-6} \text{ T} \) in \( \mu_0 H \). However, the effective lifetime is significantly altered by mul-

![Fig. 1. Observed modulation functions at \( \omega_0 \): (a) symmetric function; (b) antisymmetric function. (Arbitrary base lines.) Polarizer and analyser both set for RH circularly polarized light. \( \mu_0 H_1 = 3.8 \times 10^{-6} \text{ T} \).]
multiple scattering and collision broadening. In addition, there appears to be some cause of broadening not directly attributable to these causes; for instance, Chéron [7] observed that the Hanle curves for Hg-Cd sensitized fluorescence were significantly broader than those for Cd resonance fluorescence.

Moreover, because of the uncertainty in the effective $\Gamma_B$, the exact value of $b/\Gamma_B(=\gamma H_1/\Gamma_B)$ is difficult to ascertain. For the $\omega_0$ and $2\omega_0$ experiments, with $\Gamma_B = 444 \times 10^5$ s$^{-1}$, $b/\Gamma_B = 1.13$ and 2.25 respectively, but it appears likely that the effective values are somewhat smaller. In figure 2, the Bloch functions are plotted for $b/\Gamma_B = 0.5$ and 1, and in figure 3b we plot curves for $b/\Gamma_B = 1$ and 2.

One important point has still to be considered. The signal shapes have been calculated on the assumption that the collision axes are isotropically distributed, and that each collision gives the same contribution to $I_s(t)$ regardless of its axis. This will not in fact be true if there is a significant amount of multiple scattering in the Cd [8].

Accordingly, the contribution to $I_s(t)$ has been calculated for a single collision [8] from which the signal shapes have been deduced for the experiments reported here. We find that:

a) the $\omega_0$ signal shapes for a single collision are exactly the same as the Bloch functions in eq. (2). Multiple scattering in the Cd introduces no new complications in the expected signals. The observed signals are seen to be in reasonably good agreement with them (the Bloch functions). This is in contrast to the observations for modulation at $\omega_0$ in alignment.
transfer [2, 3] where there is disagreement between experiment and theory, indicating a significant amount of multiple scattering in the Cd and/or the Hg [8].

b) for the 2 co signals, the ratio of admixture of the functions $b^2/(b^2 + b^2 + \Gamma^2)$ and $E$ in (3) depends on the collision axis. The observed signal should therefore also be an admixture, but the ratio of the two components is difficult to ascertain. A full analysis indicates that the $E$ component is somewhat enhanced. The signal to noise ratio of the observed signals is too poor for strong conclusions to be drawn, but the general shape of the curve is much as expected, especially with regard to the displacement of the peak towards a negative value of $H$.

We note too that the incident Hg radiation undergoes multiple scattering. Evans and Gough [8] supposed that the effect of this was to enhance the population component ($T^{(0)}_d$). It can be shown that there is no effect on the shapes of the signals reported.

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