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The transfer of coherence by collisions of ^{3}He atoms

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Abstract. Magnetic resonance in the ground states of ^{3}He has been studied through the interaction with metastable atoms in a gas discharge. Modulation showing the characteristics of the ground-state resonances is observed in a transverse beam of light absorbed by the metastable atoms. This is evidence of the transfer of transverse magnetization (coherence between eigenstates) by collision. A theory is developed which explains the observations in detail.

The fact that coherence can be transferred by spin exchange in collision offers the possibility of exploitation in level-crossing or modulation experiments on spectroscopically inaccessible systems.

1. Introduction

In a type of optical pumping experiment first performed by Dehmelt (1957) a mixture of vapours is illuminated by the polarized resonance radiation of one of them, and polarization so generated in this system is communicated to the other by collisions. Magnetic resonance experiments in the second system may be monitored by changes in transparency of the first.

By an extension of the method nuclear resonances in the ground state of ^{3}He have been studied (Colegrove *et al.* 1963, Greenhow 1964). The interacting systems in this case are metastable ^{3}He atoms (1s2s $^3S_1$) polarized by optical pumping as described in the preceding paper (Partridge and Series 1966), and ^{3}He atoms in ground states (1s^2 $^1S_0$). Since, in the ground states, the electronic angular momentum is zero, the polarization which the atoms acquire by collision is entirely nuclear. Magnetic resonance at the nuclear precession frequency can be monitored by studying the absorption of radiation (2 ^{3}S–2 ^{3}P:10 830 Å) by the metastable atoms.

In experiments of this type attention has usually been directed to the longitudinal polarization. It is known that the transfer of longitudinal polarization is very efficient. The cross section for this process in ^{3}He is of the order of 4×10^{-16} cm^2. Less attention has been paid to the transfer of transverse polarization, although Colegrove *et al.*, and also Greenhow, monitored the transverse relaxation of ^{3}He nuclei with a transverse beam of light. Schearer *et al.* (1963) also observed modulation at the nuclear resonance frequency in the transverse beam and constructed a magnetometer based on their observations. The experiments reported here were designed to extend the observations and interpret the phenomenon.

Ruff and Carver (1965) have recently performed similar experiments with the Na–H system, both types of atom being in the ground states. Modulation at the hydrogen resonance frequency was observed in a transverse beam of sodium light.

We wish to underline the point which Ruff and Carver make concerning the significance of experiments of this type. Observation of modulation is interpreted as

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evidence of coherence between the eigenstates (that is, transverse polarization) of the absorbing system. This coherence, the characteristics of which are those of the system undergoing resonance, must have been transferred to the absorbing system in the process of collision. If coherence can be transferred in this way, then it should be possible to apply the recently developed spectroscopic techniques which depend on coherence between eigenstates to systems which themselves are spectroscopically inaccessible. The point should not be overlooked, however, that the success of Ruff and Carver's experiment and of our own depends on the strength of the electron exchange in relation to other interactions.

In §§ 2 and 3 of this paper we shall describe the experiments and the observations, and in § 4 develop a theory in terms of which the observations may be interpreted. The theory is based on earlier theories of spin exchange (Wittke and Dicke 1956, Purcell and Field 1956, Balling *et al.* 1964), and incorporates the concept of metastability exchange (Colegrove *et al.* 1963). A 'strong' collision between one atom in the ground state and one in the metastable state results in an exchange of electron spins and excitation energy, so that a nucleus which enters the collision in a ground-state atom may leave it in a metastable atom. It is assumed here that the transverse, as well as the longitudinal, components of the spins are conserved in the collision. Owing to the shortness of duration of the collision in relation to the hyperfine interaction the nuclear and electron spins in the newly formed metastable atom are entirely uncorrelated. However, because the spin orientations of the nuclei which enter the metastable atoms are conserved, a precessional motion at the driving frequency of the nuclear resonance is transmitted to these atoms. This frequency is very different from their Larmor frequency. The amplitude of the response is determined by the difference between the two frequencies in relation to the damping constant.

The principles of the theory could be applied to other colliding spin systems, but the details in § 4 are worked out for the particular system under discussion.

2. The experimental arrangement

This closely resembled the arrangement described in the preceding paper (Partridge and Series 1966). It is shown diagrammatically in figure 1.

The sample cell in this experiment contained ^{3}He gas at a pressure of 1 mmHg. The same cell was used by Greenhow (1964) for his experiments on nuclear nutation in ^{3}He.

The sample was pumped by circularly polarized 1 μm radiation from a ^{4}He lamp. The cell was placed in a weak static field H, of order 0·2–0·4 G. The radio-frequency field for magnetic resonance (H_1) was of amplitude less than 1 mG, at a frequency 1·07 kc/s ($\omega_0/2\pi$) in the perpendicular plane. Since this frequency is three orders of magnitude larger than the resonance linewidths, an oscillating field was used, and the perturbation due to the counter-rotating component ignored.

The monitoring lamp used in the cross beam contained ^{3}He at a pressure of 1·5 mmHg. It was constructed with a re-entrant window to reduce self-reversal of the 1 μm radiation.

The detecting equipment allowed phase-sensitive detection of the modulated photoelectric signal, rectification, and direct recording.

3. Experimental results

The experiments reported by Schearer *et al.* (1963) were first repeated. It was confirmed that modulated absorption was present in the cross beam at those values of

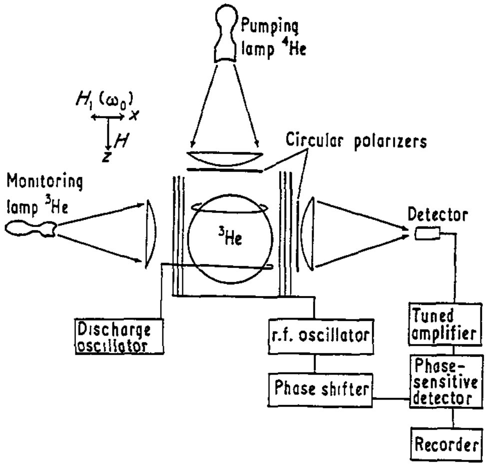

Figure 1. Disposition of apparatus. The static magnetic field H was in the direction of the pumping beam. The radio-frequency field H_1 was in the direction of the monitoring beam.

the magnetic field which satisfied the condition for *nuclear* resonance at the applied frequency. The experiments were then extended in an attempt to confirm some of the predictions arising from the theory presented in § 4.

3.1. *Resonance functions*

The modulation was present when the sample was monitored by circularly polarized light, as in the experiments of Bell and Bloom (1957). The symmetrical resonance signal found by Schearer *et al.* was accompanied by an antisymmetrical signal in quadrature as predicted by the theory (equation (19*b*)). Representing the modulated part of the signal by

$$I_A = \chi''_{\text{exp}} \cos \omega_0 t - \chi'_{\text{exp}} \sin \omega_0 t$$

we may compare χ''_{exp} and χ'_{exp} with the corresponding functions derived from the theory. These are the familiar Bloch (1946) functions:

$$\chi' = \frac{b\delta}{\delta^2 + b^2 + \Gamma_g{}^2}, \qquad \chi'' = \frac{b\Gamma_g}{\delta^2 + b^2 + \Gamma_g{}^2} \tag{1}$$

with $b = \gamma_g H_1$, $\delta = \gamma_g(H - H_0)$, and $H_0 = \omega_0/\gamma_g$. Γ_g is the damping constant and γ_g the gyromagnetic ratio. The subscript g indicates that Γ_g and γ_g refer to the ground states, not the metastable states.

The experimental and theoretical functions are compared in figure 2 (see p. 986). The qualitative agreement is entirely satisfactory.

The dependence of χ'' and χ' on b was tested. The linewidth proved to be sensitive to spatial inhomogeneities in b, but with a sufficiently homogeneous field, the dependence predicted by (1) was verified.

3.2. *Damping constant*

Measuremenf of $\Delta_{1/2}$, the half-width of the χ'' curves at half-height, allowed an experimental determination of the damping constant by use of the relation

$$\Gamma_g^2 = \Delta_{1/2}^2 - (\gamma_g H_1)^2 \tag{2}$$

with $\gamma_g = 3{\cdot}245$ kc/s G^{-1} (Anderson 1949).

The value of Γ_g so obtained depended on the discharge conditions in the cell: values ranging from 1·0 to 3·0 c/s were found. These values agree well with those obtained by Greenhow (1964) using the same sample cell, but a different experimental method. They are larger than the values reported by Schearer *et al.* (1963).

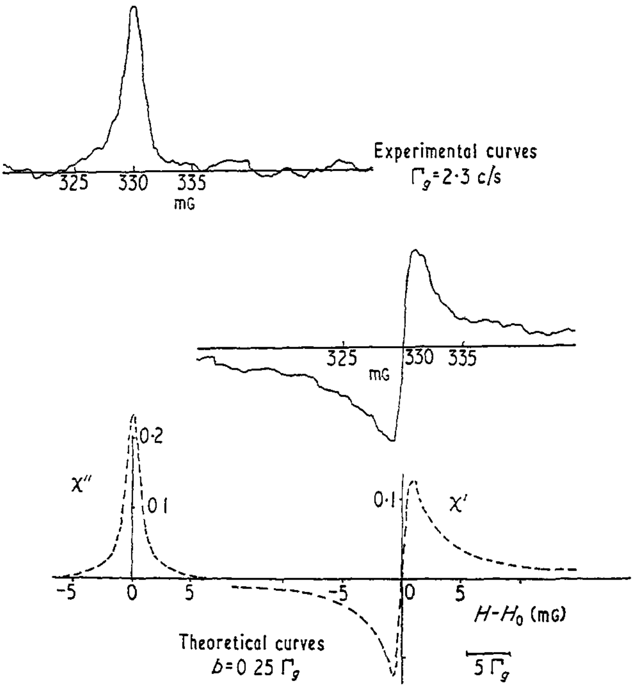

Figure 2. The amplitudes of modulation, χ' and χ'', as functions of magnetic field H. For the experimental curves, $H_1 = 0{\cdot}18$ mG, $\omega_0/2\pi = 1{\cdot}07$ kc/s, $\Gamma_g = 2\ 3$ c/s. The integrating time constant was 1 sec. The theoretical curves were plotted for $b/\Gamma_g = 0{\cdot}25$, corresponding to the experimental conditions.

It is particularly to be noticed that these resonance curves, of widths a few cycles per sec, were studied by monitoring atoms, the lifetimes of which are of the order of 10^{-4} sec.

3.3. *Polarization of the light*

The results quoted above were obtained by the use of a circular polarizer in the cross beam, either before or after the sample cell. With a linear polarizer the signals disappeared almost entirely. For no orientation of the polarizer, placed before or after the cell, was modulation found in excess of 1% of the effect with a circular polarizer.

The theory predicts that no modulation should be generated with a linear polarizer. We interpret the small signals as arising from small departures from the ideal geometrical conditions, and regard the experimental test as a confirmation of the theory.

We attach as much importance to this null result as to the positive results obtained with a circular polarizer. For it is predicted that any system having spin greater than $\frac{1}{2}$ should yield, with this geometrical arrangement, modulation signals represented by the B and C functions mentioned in the preceding paper, or some linear combination of them (Carver and Partridge 1966). The hyperfine structure of the $2\,^3S_1$ states has $F = \frac{1}{2}$ and $\frac{3}{2}$. The fact that strong modulation was not found in the cross beam using a linear polarizer is further evidence that the modulation effects are being generated in a spin $\frac{1}{2}$ system, rather than in the metastable atoms themselves.

4. Theoretical analysis

Our aim will be to determine the effect of collisions on the density matrix for atoms in metastable states when the colliding atoms are in ground states undergoing magnetic resonance. With knowledge of this density matrix it is a straightforward, though tedious, matter to calculate the absorption of light.

4.1. *The notation*

Let $\sigma(g)$ denote the partial density matrix for atoms in the ground states $1s^2\,^1S_0$. In these states there is no hyperfine interaction, and the conventional labels (F, m_F) are identical with (I, m_I). g will be used to label the states, and to represent the value of m_I. The axis of quantization is in the direction of the static field H.

Let $\sigma(\mu)$ denote the partial density matrix for atoms in the metastable states $1s2s\,^3S_1$. μ will be used to label the hyperfine states (F, m_F), and to represent the value of m_F. We shall need to express $\sigma(\mu)$ also in the decoupled representation $\sigma^*(m_I, m_J)$. (The asterisk serves to identify the matrix as describing the metastable atoms.) Let T be the transformation matrix, so that

$$\sigma(\mu) = T\sigma^*(m_I, m_J)T^{-1}. \tag{3}$$

The electronic properties of the metastable atoms are described by the density matrix $\sigma^*(e)$, the elements of which are

$$\sigma^*(m_J, m_J') = \sum_{m_I} \sigma^*(m_I, m_J; m_I', m_J')\delta(m_I, m_I'). \tag{4a}$$

The nuclear properties are described by the matrix $\sigma^*(n)$, the elements of which are

$$\sigma^*(m_I, m_I') = \sum_{m_J} \sigma^*(m_I, m_J; m_I', m_J')\delta(m_J, m_J'). \tag{4b}$$

4.2. *The collisions*

$\sigma(g)$ and $\sigma(\mu)$ and its contracted forms describe the steady-state properties of the assembly. The collisions introduce, on the one hand, loss, and on the other hand, regeneration, for both ground and metastable atoms. If we represent the collisions as a sequence of uncorrelated processes occurring at a uniform rate, we may describe the loss and regeneration by introducing rate constants.

Each collision will yield a pair of atoms, of which one is in the ground state and one in the metastable state. There will be two types of collision, one in which the atoms (labelled by the nuclei) exchange metastability, and one in which they do not. The former case is our main interest. The latter case should, strictly, be written into the equations, but since we shall solve them by successive approximation, and since the uninteresting case does not yield a major term in the equations, we shall ignore it.

In the case where the atoms exchange metastability, the density matrices which we use to represent the products of the collision are based on the assumptions (i) that the nuclear and electronic parts separately of the density matrices are unaltered by the collisions, and (ii) that the nuclear and electronic parts of the newly formed metastable atoms are entirely uncorrelated. Accordingly, the density matrices for the ground-state and metastable atoms immediately after the collision are written $\sigma^*(n)$ and $[\sigma^*(e)\times\sigma(g)]$, respectively.

Introducing now the damping constants Γ_g' and Γ_μ' to represent loss from the ground and metastable states, respectively, and the rate constants R_g and R_m to represent regeneration, we may write differential equations for the effect of collisions. The equations are

$$\left[\frac{d}{dt}\sigma(g)\right]_{\text{coll}} = -\Gamma_g'\sigma(g)+R_g\sigma^*(n) \tag{5a}$$

$$\left[\frac{d}{dt}\sigma(\mu)\right]_{\text{coll}} = -\Gamma_\mu'\sigma(\mu)+R_m[T\sigma^*(e)\times\sigma(g)T^{-1}]. \tag{5b}$$

We shall later make use of a selection rule which can be derived from the last term of (5*b*). Since $\mu = g+m_J$, we have

$$(\mu-\mu') = (g-g')+(m_J-m_J'). \tag{6}$$

While this rule must hold in general, we wish to apply it when the matrix $\sigma^*(e)$ corresponds to a random, isotropic distribution of electron spins (the zero-order solution $\sigma^{*(0)}(e)$, § 4.4.1). In this case, the off-diagonal components of $\sigma^*(e)$ are zero, and all components of $\sigma(\mu)$ vanish unless $m_J = m_J'$. We have, therefore,

$$\mu-\mu' = g-g' \tag{6a}$$

which must be satisfied for all collisions in which the newly formed metastable atoms are described by $\sigma^{*(0)}(e)\times\sigma(g)$.

It is worth noticing that (6*a*) holds also if the electron spins are polarized but uncorrelated, for it depends, not on the equality of the diagonal elements of $\sigma^*(e)$, but on the absence of off-diagonal elements. (6) and (6*a*) are analogous to the rules which govern the transfer of coherence in the interaction of atoms with light (see Series 1966, to be referred to as I, and references quoted there).

4.3. *The equations of motion*

The complete equations for the time derivatives are obtained by including the other perturbations (static field, radio-frequency field, optical pumping, other causes of damping). The equations are

$$\frac{d}{dt}\sigma(g) = -\frac{i}{\hbar}[(\mathscr{H}_0+\mathscr{H}_{\text{stat}}+\mathscr{H}_{\text{rf}}),\ \sigma(g)]-\Gamma_g\sigma(g)+R_g\sigma^*(n) \tag{7a}$$

$$\begin{aligned}\frac{d}{dt}\sigma(\mu) = &-\frac{i}{\hbar}[(\mathscr{H}_0+\mathscr{H}_{\text{stat}}),\ \sigma(\mu)]-\Gamma_\mu\sigma(\mu)+R\mathbf{1}+B\sigma(\mu)B^+\\ &+R_m[T\sigma^*(e)\times\sigma(g)T^{-1}].\end{aligned} \tag{7b}$$

The notation is similar to that used in I, the first paper of this series. B is the operator which represents one cycle of optical pumping, and Γ_μ is taken to include all forms of damping of metastable atoms. The term $R\mathbf{1}$ represents the regeneration of atoms by the discharge at the rate R/s into a statistical ensemble of s equally populated, uncorrelated, metastable states. No regeneration term other than $R_g\sigma^*(n)$ is written for $\sigma(g)$, and Γ_g is identified with Γ_g', since the collisions constitute the major source of regeneration and damping for atoms in ground states. $\mathscr{H}_{\mathrm{rf}}$ does not appear in the equation for the $|\mu\rangle$ since the oscillating field is too weak, and too far from resonance to have any direct effect on atoms in the metastable states.

These equations can be solved by successive approximation. A zero-order solution $\sigma^{(0)}(\mu)$ may be obtained by taking the right-hand side of (7*b*) as far as the term in R. Including next the term in B, one may obtain $\sigma^{(1)}(\mu)$, a first-order increment to $\sigma^{(0)}(\mu)$. Using this in (7*a*) a solution $\sigma(g)$ may be found which, when used in the final term of (7*b*) will yield a second-order solution $\sigma^{(2)}(\mu)$. This is the contribution to $\sigma(\mu)$ which we are seeking.

4.4. *Solution of the equations*

4.4.1. $\sigma^{(0)}(\mu)$. The commutator bracket in equation (7*b*) is easily reduced to

$$-i(k_\mu - k_{\mu'})\sigma_{\mu\mu'}$$

where k_μ is the Bohr frequency of the state $|\mu\rangle$. Taking the terms in Γ_μ and R, together with the commutator bracket, the solution is

$$\sigma_{\mu\mu'}{}^{(0)}(t) = \sigma_{\mu\mu'}{}^{(0)}(0)\exp[-\{\Gamma_\mu + i(k_\mu - k_{\mu'})\}t] + \frac{R}{\Gamma_\mu}\delta_{\mu\mu'}. \tag{8}$$

The transient, as well as the steady-state, solution has been written here, since we need to know the time-development operator for the solutions below.

4.4.2. $\sigma^{(1)}(\mu)$. Proceeding as in I, § 2.4, the first-order increment, which represents the result of one cycle of optical pumping, is

$$\sigma_{\mu\mu'}{}^{(1)}(t) = \sum_{\mu_1\mu_1'} B^{\mu\mu'}_{\mu_1\mu_1'} \frac{(R/\Gamma_\mu)\delta(\mu_1,\mu_1')}{\Gamma_\mu + i(k_\mu - k_{\mu'})}. \tag{9}$$

This is the steady-state solution.

The magnitude of the off-diagonal, relative to the diagonal components of $\sigma^{(1)}(\mu)$ depends on the B coefficient, and on the magnitude of $k_\mu - k_{\mu'}$ relative to Γ_μ.

For the particular states $|\mu\rangle$ with which we are concerned, the hyperfine structure is much larger than the natural width, and the off-diagonal elements connecting states of different F will be negligibly small. Matrix elements of this sort will be discarded. On the other hand, off-diagonal elements which connect states of the same F but different m_F will not necessarily be small. For such elements, we shall write

$$k_\mu - k_{\mu'} = (\mu - \mu')g_F\omega_L$$

where $g_F\omega_L$ is the Larmor frequency of the level F, and μ, μ' are the values of m_F, m_F'.

Although we shall need these matrix elements later, we shall discard them at this stage because, if the pumping light is polarized so as to generate maximum polarization in the metastable states, the coefficient B will be zero for these off-diagonal elements. It is nevertheless worth noticing that, if B does not vanish, then the condition $\omega_L \ggg \Gamma_\mu$ (see equation (12) below), which allows the coherence in collisions to survive in the steady state, would also allow off-diagonal components of $\sigma(\mu)$ to be generated in the optical pumping cycle. These in turn would generate an initial coherence in $\sigma(g)$, and lead to modulation terms additional to those calculated below. Of these terms, those at the frequency ω_0 would be of comparable strength with those calculated; terms at harmonic frequencies would also be found, the amplitudes of which would depend on the ratio ω_L/Γ_μ.

4.4.3. $\sigma(g)$. With the exclusion of the off-diagonal elements of $\sigma^{(1)}(\mu)$, equation (9) represents a polarized incoherent assembly of metastable atoms, and $\sigma^*(n)$ is proportional to $\begin{pmatrix}1 & 0\\ 0 & 0\end{pmatrix}$. We need not write down the coefficient. The solution of (7*a*) is now straightforward (equation (8) of I, for example, without the summation over μ_0), and yields

$$\sigma_{gg'}(t) \propto \exp\{-i(g-g')\omega_0 t\} \sum_{l,l'} \frac{\langle g_0|l\rangle\langle l|g\rangle\langle g'|l'\rangle\langle l'|g_0\rangle}{\Gamma_g+i(l-l')p} \tag{10}$$

where $\langle g|l\rangle$ etc. are elements of the rotation matrix, and $p = \gamma_g\{(H-H_0)^2+H_1^2\}^{1/2}$.

4.4.4. $\sigma^{(2)}(\mu)$. Returning to (7*b*) with the expression for $\sigma(g, t)$, and using for $\sigma^*(e)$ the steady-state zero-order solution $\sigma^{*(0)}(e)$, we may integrate the equation by the methods used before. It is found that the (μ, μ', g, g') component has the denominator

$$\Gamma_\mu + i(\mu-\mu')g_F\omega_L - i(g-g')\omega_0. \tag{11}$$

The selection rule $\mu-\mu' = g-g'$, equation (6*a*), is applicable to this case. Hence, $g-g'$ may be eliminated from (11) in favour of $\mu-\mu'$, and the solution of equation (7*b*) written in the simple form

$$\sigma_{\mu\mu'}^{(2)}(t) \propto [T\sigma^{(0)}(e)\times\sigma(g,t)T^{-1}]_{\mu\mu'} \frac{R_m}{\Gamma_\mu+i(\mu-\mu')(g_F\omega_L-\omega_0)}. \tag{12}$$

This result shows how the time dependence of $\sigma(g, t)$ is incorporated into $\sigma^{(2)}(\mu, t)$. It goes beyond the assumptions concerning the collisions in that it describes the steady-state situation, rather than the effect of a single pulse. Assumption (i) was that all the components of $\sigma(n)$ are transferred in the collision, whereas (12) shows that, in the steady state which results from a sequence of uncorrelated pulses at a uniform rate, the off-diagonal components of the density matrix do not survive if $g_F\omega_L-\omega_0 \gg \Gamma_\mu$. A condition of this sort is not peculiar to the collision interaction: it is a feature of rate processes in general, and in particular of the optical pumping cycle (cf. § 4.4.2).

A diagrammatic representation of the condition for the survival of coherence is illustrated in figure 3. k is proportional to the energy exchanged in the collision. An

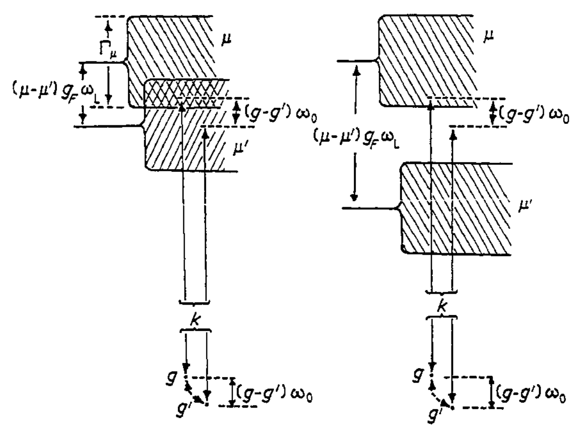

Figure 3. Condition for the survival of coherence in collisions. k is proportional to the energy exchanged in the collision. Case (*a*), $(\mu-\mu')g_F\omega_L-(g-g')\omega_0 < \Gamma_\mu$, k falls within the region of resonance for μ and μ' and coherence survives; case (*b*), $(\mu-\mu')g_F\omega_L-(g-g')\omega_0 > \Gamma_\mu$, k falls within the region of resonance for μ, but not for μ', and coherence does not survive.

arrow labelled k links the states $|g\rangle$ and $|\mu\rangle$ of the atom which is to be excited. An arrow of the same length links $|g'\rangle$ and $|\mu'\rangle$ also. The diagram shows that if k occurs within the resonance region for one transition, and if

$$(\mu-\mu')g_F\omega_L-(g-g')\omega_0 < \Gamma_\mu$$

then both transitions may be stimulated, whereas if $(\mu-\mu')g_F\omega_L-(g-g')\omega_0 > \Gamma_\mu$, one transition or the other, but not both, may be stimulated. Coherence which may have existed between $|g\rangle$ and $|g'\rangle$ will be transferred to $|\mu\rangle$ and $|\mu'\rangle$ in the first case, but not in the second. The condition simplifies to the form given above by use of the selection rule $\mu-\mu' = g-g'$, with $g, g' = \pm\frac{1}{2}$.

4.5. *Explicit form of* $\sigma^{(2)}(\mu, t)$

The transformation matrix T consists of the array of Wigner coupling coefficients as shown in the table.

F, m_F \\ m_J, m_I	$1, \frac{1}{2}$	$1, -\frac{1}{2}$	$0, \frac{1}{2}$	$0, -\frac{1}{2}$	$-1, \frac{1}{2}$	$-1, -\frac{1}{2}$
$\frac{3}{2}, \frac{3}{2}$	1	0	0	0	0	0
$\frac{3}{2}, \frac{1}{2}$	0	$(\frac{1}{3})^{1/2}$	$(\frac{2}{3})^{1/2}$	0	0	0
$\frac{3}{2}, -\frac{1}{2}$	0	0	0	$(\frac{2}{3})^{1/2}$	$(\frac{1}{3})^{1/2}$	0
$\frac{3}{2}, -\frac{3}{2}$	0	0	0	0	0	1
$\frac{1}{2}, \frac{1}{2}$	0	$(\frac{2}{3})^{1/2}$	$-(\frac{1}{3})^{1/2}$	0	0	0
$\frac{1}{2}, -\frac{1}{2}$	0	0	0	$(\frac{1}{3})^{1/2}$	$-(\frac{2}{3})^{1/2}$	0

We have also

$$\sigma^{*(0)}(e) \equiv \sigma^{*(0)}(m_J, m_J') = \tfrac{1}{3}\begin{pmatrix}1 & 0 & 0\\ 0 & 1 & 0\\ 0 & 0 & 1\end{pmatrix} \tag{13}$$

and we shall write

$$\sigma(g, t) = \begin{pmatrix}\sigma_{++} & \sigma_{+-}\\ \sigma_{-+} & \sigma_{--}\end{pmatrix}. \tag{14}$$

The matrix elements are given by equation (10), in which g, g', l, l', take the values $\pm\frac{1}{2}$. In writing the denominators $\Gamma_\mu+i(\mu-\mu')(g_F\omega_L-\omega_0)$, we shall suppress the subscript on Γ_μ, and write $g_F\omega_L-\omega_0$ as ω_a, ω_b for $F = \frac{3}{2}$ and $F = \frac{1}{2}$ respectively.

Using these expressions in equation (12) we find

$$[\sigma^{(2)}(\mu, t)]_{F=3/2} = A\begin{pmatrix}\dfrac{3\sigma_{++}}{\Gamma} & \dfrac{3^{1/2}\sigma_{+-}}{\Gamma+i\omega_a} & 0 & 0\\ \dfrac{3^{1/2}\sigma_{-+}}{\Gamma-i\omega_a} & \dfrac{2\sigma_{++}+\sigma_{--}}{\Gamma} & \dfrac{2\sigma_{+-}}{\Gamma+i\omega_a} & 0\\ 0 & \dfrac{2\sigma_{-+}}{\Gamma-i\omega_a} & \dfrac{\sigma_{++}+2\sigma_{--}}{\Gamma} & \dfrac{3^{1/2}\sigma_{+-}}{\Gamma+i\omega_a}\\ 0 & 0 & \dfrac{3^{1/2}\sigma_{-+}}{\Gamma-i\omega_a} & \dfrac{3\sigma_{--}}{\Gamma}\end{pmatrix} \tag{15a}$$

and

$$[\sigma^{(2)}(\mu, t)]_{F=1/2} = A\begin{pmatrix}\dfrac{\sigma_{++}+2\sigma_{--}}{\Gamma} & \dfrac{-\sigma_{+-}}{\Gamma+i\omega_b}\\ \dfrac{-\sigma_{-+}}{\Gamma-i\omega_b} & \dfrac{2\sigma_{++}+\sigma_{--}}{\Gamma}\end{pmatrix} \tag{15b}$$

where A is a constant.

4.6. *Cross beam modulation signals*

4.6.1. *Spectral density of the light.* The rate of absorption of radiation by the metastable atoms is given by the generalized form of I, equation (14):

$$L_{\mathrm{A}} = \mathrm{Tr}[\mathscr{A}\sigma(\mu, t)] \tag{16}$$

where $\mathscr{A}_{\mu\mu'} = \Sigma_m(k_m/\hbar)\rho(k_m)\langle\mu'|\mathbf{e}_i^{0*}.\mathbf{P}|m\rangle\langle m|\mathbf{e}_i^0.\mathbf{P}|\mu\rangle$, $\mathbf{e}_i^0$ is the unit vector specifying the polarization of the light, $\mathbf{P}$ is the electric dipole operator, and the $|m\rangle$ are the states belonging to 2 $^3\mathrm{P}_{0,1,2}$. $\rho(k_m)$ is the spectral density of the light in the region of absorption, k_m.

If $\rho(k_m)$ were constant for all transitions in the sum over m, the net absorption would be constant. This is because of the orbital spherical symmetry of the metastable level. For the lamps used in the investigation (both ^{3}He and ^{4}He) the spectral density was not constant over the m.

4.6.2. *Character of the polarizer.* A significant difference between the results to be expected in monitoring magnetic resonance experiments with a linear and with a circular polarizer was pointed out by Carver and Partridge (1966). The present case affords an example.

Evaluation of the monitoring operator $\mathscr{A}$ for the linear polarizer specified by the vector $\mathbf{e}^0 = \mathbf{k}\sin\theta + \mathbf{j}\cos\theta$ yields, for the states $F = \frac{1}{2}$,

$$\mathscr{A}^{\mathrm{lin}}_{F=1/2} = K\begin{pmatrix} 1+K'\cos^2\theta & 0 \\ 0 & 1+K'\cos^2\theta \end{pmatrix} \tag{17}$$

where K and K' are constants which depend on the spectral density of the light. Formation of the trace specified in equation (16) shows that only the diagonal components of $\sigma(g, t)$ appear in the result; that is to say, the $F = \frac{1}{2}$ components will contribute no modulated absorption to the cross beam. Similarly it may be shown that no modulation is contributed by the $F = \frac{3}{2}$ components. It is predicted that the absorption from a linearly polarized beam should be unmodulated.

This result is characteristic of systems having spin $\frac{1}{2}$, and derives from the spin $\frac{1}{2}$ system out of which $\sigma(\mu, t)$ was built. The result does not apply to systems having spin greater than $\frac{1}{2}$. Modulation would have been found for a linearly polarized cross beam monitoring magnetic resonance within the states of $F = \frac{3}{2}$ themselves.

On the other hand, the monitoring operator which corresponds to the circular polarizer specified by $\mathbf{e}^0 = 2^{-1/2}(\mathbf{k}+i\mathbf{j})$ is, for the states $F = \frac{1}{2}$,

$$\mathscr{A}^{\mathrm{circ}}_{F=1/2} = \begin{pmatrix} K'' & K''' \\ K''' & K'' \end{pmatrix} \tag{18}$$

where K'' and K''' again are constants. Formation of the trace in (16) now leads to the result

$$(L_{\mathrm{A}})_{F=1/2} = \frac{\mathrm{const.}}{\Gamma^2+\omega_{\mathrm{b}}^2}\left\{\mathrm{const.} + \frac{b(\Gamma\delta+\Gamma_g\omega_{\mathrm{b}})}{\Gamma_g(\delta^2+b^2+\Gamma_g^2)}\cos\omega_0 t + \frac{b(\Gamma\Gamma_g-\omega_{\mathrm{b}}\delta)}{\Gamma_g(\delta^2+b^2+\Gamma_g^2)}\sin\omega_0 t\right\} \tag{19}$$

and a similar expression for $(L_{\mathrm{A}})_{F=3/2}$.

This is the modulation in which we are interested. The amplitudes of modulation are resonance functions of the variable δ with the characteristics of magnetic resonance in the ground states.

It is instructive to study equation (19) in the limiting cases $\omega_{\mathrm{b}} \ll \Gamma$ and $\omega_{\mathrm{b}} \gg \Gamma$. Although we are thinking of δ as the variable, its value in the region of resonance will be of the order of Γ_g. We have, therefore, the following cases:

case (*a*) $\omega_b \ll \Gamma$

$$(L_A)_a \rightarrow \frac{\text{const.}}{\Gamma\Gamma_g}\left(\text{const.}+\frac{b\delta}{\delta^2+b^2+\Gamma_g^2}\cos\omega_0 t+\frac{b\Gamma_g}{\delta^2+b^2+\Gamma_g^2}\sin\omega_0 t\right); \quad (19a)$$

case (*b*) $\omega_b \gg \Gamma$

$$(L_A)_b \rightarrow \frac{\text{const.}}{\omega_b\Gamma_g}\left(\text{const.}+\frac{b\Gamma_g}{\delta^2+b^2+\Gamma_g^2}\cos\omega_0 t-\frac{b\delta}{\delta^2+b^2+\Gamma_g^2}\sin\omega_0 t\right). \quad (19b)$$

In case (*b*) the signal is smaller than in case (*a*) by the factor ω_b/Γ. This exemplifies the condition for the survival of coherence, $\omega_b \ll \Gamma$.

For reasons of practical convenience this condition was not satisfied in the experiments. Numerical values were: $\Gamma \sim 75$ kc/s; $\omega_b \sim 600$ kc/s. The fact that signals were detected under these unfavourable conditions demonstrates the efficiency of the postulated mechanism for the transfer of coherence in collisions.

5. Conclusion

It has been confirmed that transverse magnetization in the ground states of ^{3}He leads to modulation in light absorbed by metastable atoms. Earlier studies (Schearer *et al.* 1963) emphasized the application to magnetometry. The present interpretation of these experiments in terms of the transfer of coherence between eigenstates suggests that the spectroscopic techniques which rely on such coherence (modulation and level-crossing phenomena) might be applicable to systems which are themselves spectroscopically inaccessible. It is unlikely, however, that the transfer of coherence would be efficient if interactions other than electron exchange dominated the collisions, or if the frequency mismatch of the systems were greatly in excess of the damping constant of the receiving system.

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