OPTICAL PUMPING—A FORMALISM AND PROOF

by B. S. MATHUR, *Department of Physics, Indian Institute of Technology, Kanpur 16*

A striking parallelism between the role of atoms and light in 'Optical-pumping' experiments has been discussed. It is shown that the evolution of atoms in their ground state can be discussed in terms of an effective Hamiltonian $\delta\mathcal{H}$ and similarly the propagation of light through a polarized vapour can be discussed in terms of an effective Hamiltonian $-2\pi k\chi_\perp$ for light. An optical double-quantum transition experiment has been discussed as partial proof of the optical-pumping formalism.

INTRODUCTION

There is a striking parallelism in the role of atoms and light in optical-pumping experiments. The effect of the light on the evolution of atom in the ground state can be represented by an effective Hamiltonian $\delta\mathcal{H}$ (Happer and Mathur 1967*a*) such that the ground state satisfies the wave equation

$$i\frac{\partial\psi_g}{\partial t}=(\mathcal{H}_0+\delta\mathcal{H})\psi_g. \qquad (1)$$

$\delta\mathcal{H}$ in general is not Hermitian but can be expanded into a Hermitian and an anti-Hermitian part as

$$\delta\mathcal{H}=\delta\epsilon-i\hbar(\delta\Gamma/2) \qquad (2)$$

where

$$\delta\epsilon=\frac{\delta\mathcal{H}+\delta\mathcal{H}^+}{2} \quad \text{and} \quad \delta\Gamma=\frac{i}{\hbar}(\delta\mathcal{H}-\delta\mathcal{H}^+).$$

$\delta\epsilon$ might be called a light shift operator and is due to off-resonant component of the light. $\delta\Gamma$ describes the absorption of light by the atoms and is due to the resonant component of light.

Similarly the propagation of light beam in an optically-pumped alkali vapour can be described by the following propagation equation of transverse electric amplitudes which can be derived from Maxwell's equations (Mathur *et al.* 1970),

$$i\left(\frac{\partial\vec{\epsilon}_\perp}{\partial\xi}\right)=-2\pi k\chi_\perp\vec{\epsilon}_\perp \qquad (3)$$

where electric field of quasi-monochromatic light has been taken as

$$\vec{E}=\vec{\epsilon}(\rho,t)\exp i\left(\vec{k}.\vec{r}-wt\right)+c.c.$$

$\xi=\frac{1}{2}(\rho+ct)$ gives the distance a given wave front has travelled in the direction of propagation. Another variable $\eta=\frac{1}{2}(\rho-ct)$ is held constant. $\chi_\perp$ is the projection of the susceptibility operator in the plain of the wave front.

The effective Hamiltonian operator $\delta\mathcal{H}$ and the susceptibility operator are related through polarizability operator (α) as

$$\delta\mathcal{H} = \vec{\epsilon}^{*}\alpha\vec{\epsilon}$$

$$\frac{\chi}{N} = <\psi_g \mid \alpha \mid \psi_g>$$

where N is the atomic density and ψ_g is the ground state wave function.

The polarizability operator α is a joint operator, both for the light and for the atoms. To get the effective Hamiltonian for the atoms we take the expectation value of α with respect to the light. To get the susceptibility, that is the effective Hamiltonian for the light, we take the expectation value of α with respect to the atoms. The polarizability operator can be expanded as

$$\alpha = \sum_L \alpha^L = \alpha^0 + \alpha^1 + \alpha^2 + \ldots$$

into a scalar part, a vector part, a tensor part and so on.

Study of Alkali Atoms

We shall only be interested in the study of alkali atoms at this stage. In particular, we shall study the evolution of atoms in their ground state in presence of off-resonant light. So we will only be concerned with virtual transitions.

Light shift in the alkali atoms due to virtual transition can be described by a simple light shift operator (Mathur *et al.* 1968),

$$\delta\epsilon = \delta\epsilon_0 + h\,\delta A\,\vec{I}.\vec{J} - \vec{\mu}.\delta\vec{H} + \delta\epsilon_2 . \quad .. \quad .. \quad .. \quad (4)$$

Each term in the light shift operator produces a characteristic type of energy splitting of the ground state sub-levels of the atoms (Fig. 1).

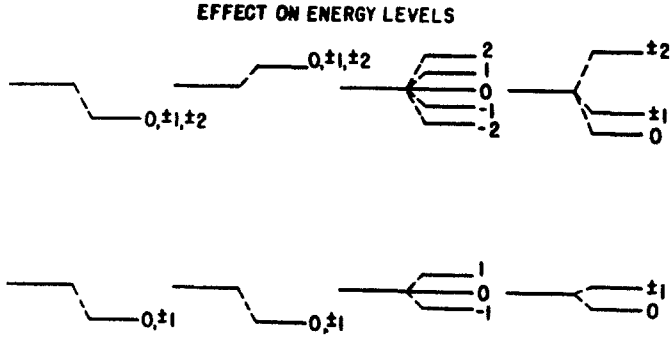

Fig. 1. The different types of light shift for an alkali atom.

The C.M. light shift, represented by $\delta\epsilon_0$, is an equal downward (or upward) displacement of all the ground state sub-levels. The hfs light shift,

which is represented by the term $h\,\delta A\,\vec{I}\,.\,\vec{J}$, is entirely equivalent to a shift in the magnetic-dipole coupling constant of the alkali-atom ground state. This will effect the frequencies of hfs transitions ($\Delta F = 1$ transitions) but not the frequencies of Zeeman transitions ($\Delta F = 0$ transitions). The Zeeman light shift operator $-\vec{\mu}\,.\,\delta\vec{H}$, which will be present only when light beam has some degree of circular polarization, can be thought of as an interaction of the magnetic dipole moment $\vec{\mu}$ of the atom with an effective magnetic field $\delta\vec{H}$ which is directed parallel or antiparallel to the light beam. Consequently, the Zeeman light shift operator will effect the Zeeman frequencies of the atom in exactly the same way as a small magnetic field $\delta\vec{H}$. The tensor shift, represented by the term $\delta\epsilon_2$, is analogous to the tensor stark shift which is caused by a static electric field.

The hfs and C.M. light shift are due to scalar part of the polarizability operator, the Zeeman light shift operator is due to vector part and tensor light shift operator is due to tensor part of the polarizability operator.

Again to show the similarity between the atoms and the light we shall write down the susceptibility dyadic (Mathur *et al.* 1970) for alkali vapour (Fig. 2),

$$\chi = \chi_{\rm eq} + \chi_{\rm hfs}\vec{I}\,.\,\vec{J} + i\sum_{ff'}\chi_{\rm gt}(ff')\vec{J}(ff')X + \sum_{ff'}\chi_{\rm br}(ff')\overleftrightarrow{Q}(ff'). \qquad (5)$$

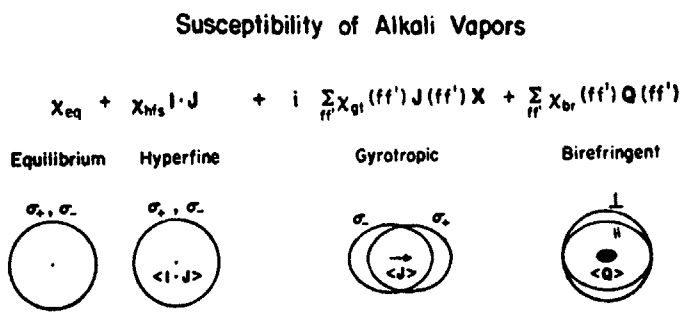

FIG. 2. Wave surfaces and the susceptibility.

Beneath each polarizability component of Fig. 2 are sketched the wave surfaces which will result if the non-equilibrium polarizability were determined by that component alone.

The equilibrium susceptibility $\chi_{\rm eq}$ is independent of the state of vapour, and it is this isotropic susceptibility which would be obtained if the atoms were distributed at random among the sub-levels of the ground state. $\chi_{\rm eq}$ is closely related to the centre-of-mass light shift operator. The hyper-fine

structure (hfs) susceptibility $\chi_{\text{hfs}}<\vec{I}.\vec{J}>$ is also isotropic, but it has a non-zero expectation value only when some population imbalance $<\vec{I}.\vec{J}>$ exists between the hyper-fine levels of the ground state. The hfs susceptibility is closely related to the hfs light shift operator. The gyrotropic susceptibility $\Sigma_{ff'}\chi_{\text{gt}}(ff')\vec{J}(ff')X$ is an anisotropic susceptibility which depends upon the expectation values $<\vec{J}(ff')>$ of the electronic angular momentum operator within each hyper-fine level ($f=f'$), and between different hfs levels ($f\neq f'$). It is closely related to the Zeeman light shift operator. The birefringent susceptibility $\Sigma_{ff'}\chi_{\text{br}}(ff')\overleftrightarrow{Q}(ff')$ is also an anisotropic susceptibility which depends on the expectation value $<\overleftrightarrow{Q}(ff')>$ of the atomic quadrupole operator.

Though we have carried out experiments to check the validity of each term of light shift ($\delta\epsilon$) and susceptibility (χ) operators of 'Optical-pumping Formalism'—here we will only be concerned with the discussion of the Zeeman light shift operator ($-\vec{\mu}.\delta\vec{H}$) part of the light shift operator.

Inverse Raman Effect

An effect of this kind, we will call an 'Optical Double-quantum Transition' experiment or an 'Inverse Raman Effect' experiment (Happer *et al.* 1967*b*). In Raman effect, when radiation at frequency ω propagates through a Raman active medium, optical double-quantum transitions take place between different initial and final vibrational levels and one observes sidebands introduced on to the light, which we call Stokes and anti-Stokes radiation. Suppose we do the experiment backwards, i.e. let time t go to $(-t)$. We will have a carrier frequency and sidebands (Stokes and anti-Stokes) going in the Raman active medium and we will end up inducing various vibrational transitions in the medium. The subject of discussion so formed is this kind of effect.

We have already said that the Zeeman light shift operator $-\vec{\mu}.\delta\vec{H}$ can be thought of as an interaction of magnetic dipole moment $\vec{\mu}$ of the atom with an effective magnetic field $\delta\vec{H}$. I will like to remind you that $\delta\vec{H}$ is not a radiation field. It is just an operator form of representing double-quantum transitions. The Zeeman light shift operator has a diagonal part and this causes small shifts (or splitting) of the energy levels of the atomic ground state and an off-diagonal part which, when light is modulated at Zeeman resonance frequency, can cause transitions between different Zeeman levels of the ground state. So if we shine a nearly resonant circularly polarized radiation on a vapour in a magnetic field *Ho*, in the same direction as *Ho*, it will amount to further splitting (positive or negative depending upon the sense of circular polarization) of the Zeeman levels or removing the degeneracy if the vapour

was originally in a zero magnetic field. Similarly it is possible to induce real transition between the Zeeman levels of the atomic ground state by illuminating the atoms in a transverse direction to the static magnetic field *Ho* with a modulated, circular polarized light. So, as far as the Zeeman coherence of atoms is concerned, the effects of the fictitious magnetic field are indistinguishable from those of a real magnetic field and in some sense, this experiment can be described as a Zeeman resonance experiment with a fictitious oscillating magnetic field.

Experimental Procedure

The basic concepts of the experiment are discussed below (Fig. 3). A beam of monochromatic light is incident on an atom which is in the sub-level i of its ground state. The frequency ω of the light is several Doppler widths removed from one of the optical absorption frequencies of the atom: When the light is amplitude modulated at the ground state transition frequency ω_{ij}, one finds that the atom makes a transition from the level i to the level j of the ground state. The amplitude modulated light contains a carrier frequency ω and two sideband frequencies $\omega \pm \omega_{ij}$. The atom absorbs a photon at the carrier frequency ω and is excited to the virtual state m. Then one of the sidebands of the modulated light stimulates the atom to re-emit a photon of frequency $\omega + \omega_{ij}$ and falls down to the state j. The frequencies are such

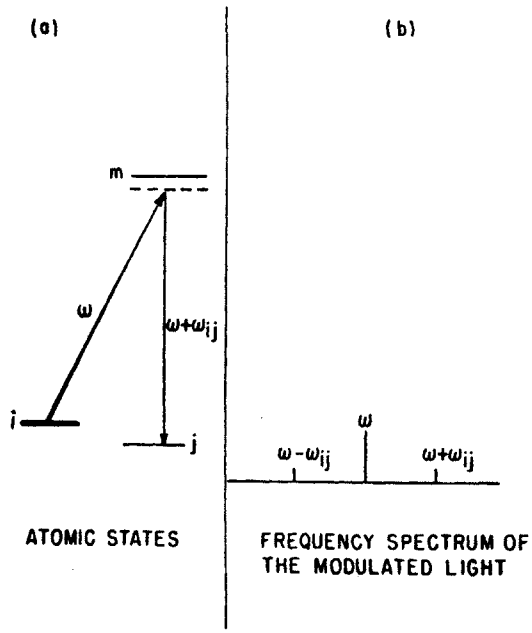

Fig. 3. Double-quantum transitions between two Zeeman levels caused by optical photons.

that energy is conserved in the process, and the polarization of the incident light must be such that angular momentum is also conserved.

The actual experiment was performed with optically pumped rubidium vapour. Since alkali atoms have a rather complicated structure in the ground state and in the excited states, there are many possible double-quantum transitions of the type illustrated in Fig. 3, involving different intermediate states m and different combinations of sideband and carrier frequencies. The amplitude of all these double-quantum processes adds coherently to produce the final transition amplitude. It can be shown that the net effect of all double-quantum transitions within the Zeeman multiplet F of the ground state can be represented by an effective Hamiltonian of the form $-\vec{\mu} \cdot \delta\vec{H}$, i.e. the Zeeman light shift operator. The fictitious magnetic field $\delta\vec{H}$ is given by

$$\delta\vec{H}(F) = \frac{\lambda^2 u}{4\pi g_J \mu_0} \frac{e^2}{mc^2} f_J \frac{\omega}{\omega - \omega_F} \left[\frac{11 - 4J(J+1)}{4}\right] \left(\frac{\vec{e} \times \vec{e}^*}{i}\right) \quad .. \quad (6)$$

where λ is the wavelength of the exciting light, and $\omega = \frac{2\pi c}{\lambda}$; f_J is the absorption oscillator strength to the ^2P_J doublet; ω_F is the average frequency between all the hfs components of ^2P_J excited state and the hfs multiplet F of the ground state. J is the electronic angular momentum of the excited ^2P_J state; g_J is the gyromagnetic ratio of the $^2S_{\frac{1}{2}}$ ground state; μ_0 is the Bohr magneton; u is the energy density of light and $\vec{e}$ is the complex polarization vector of the exciting light. $\delta\vec{H}$ will be different for different hfs levels of the ground state and also for a given light intensity and frequency displacement $\delta\vec{H}$ due to D_2 light is approximately equal and opposite to that by D_1 light. This we have checked in our experiment also.

The experimental apparatus is shown in Fig. 4. The main features of the apparatus are that a solenoid and Mumetal shields arrangement was used to reduce the field to 10 milligauss. This also reduced the field inhomogeneities and we were able to get Zeeman resonance line-width of the order of 40 Hz as observed by absorption of the pumping light. Rb^{87} vapour in a 100 ml flask was optically pumped by Rb^{87} resonance radiation and this flask was placed in the centre of the solenoid. Off-resonant light was obtained by passing Rb^{85} light through a heated Rb^{87} filter cell, thus leaving only the $F = 2$ hfs component of the Rb^{85} resonance light. The Rb^{85} light was 100 per cent square wave modulated at 10,000 Hz. $\delta\vec{H}$ obtained in our set-up was of the order of 10^{-7} gauss. If $\delta\vec{H}$ were several order of magnitude larger, it could be used to induce Zeeman resonances directly. However, for very small effective magnetic field one can obtain a considerable enhancement in signal power (10^5) by simultaneously applying a real, coherent rf magnetic field.

Thus a real 9990 Hz transverse field was also applied to the absorption flask. The strength of 9990 Hz rf was attenuated until the Zeeman resonance was half-saturated. When the static magnetic field was swept through the resonance, a typical increase in the absorption of the vapour was observed.

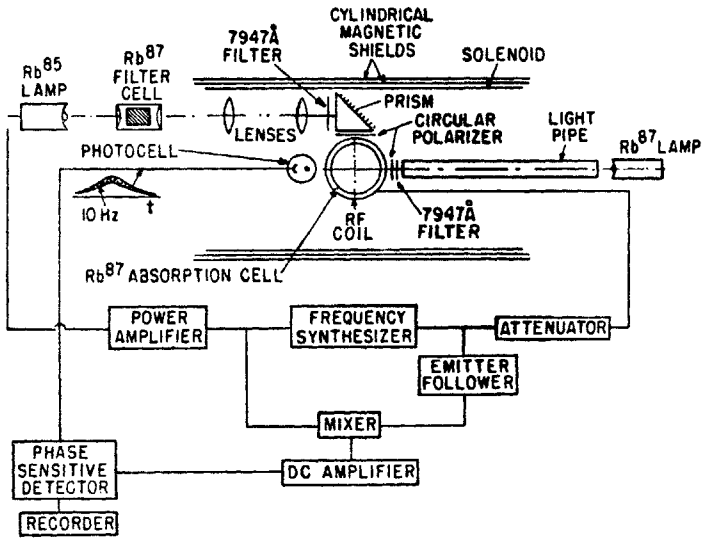

FIG. 4. The spectrometer for coherent detection of optical double-quantum transition.

However, the fictitious magnetic field due to the optical double-quantum transitions alternately increased and decreased the transition probability so that a 10 Hz ripple was present on top of the usual resonance signal. The 10 Hz ripple was detected with a phase sensitive amplifier. The 10 Hz signal disappeared when the Rb^{85} light was blocked; when the 10,000 Hz modulation was removed from the lamp, when the circular polarizer was removed from the Rb^{85} beam, when the 9990 Hz rf was absent and when the Rb^{87} pumping light was turned off. The signal amplitude and the signal-to-noise ratios were in agreement with order of magnitude theoretical estimates.

We conclude that the observed signals do indeed represent 'Optical Double-quantum Transitions' between the Zeeman sub-levels of the ground states or we can say it is an 'Inverse Raman Effect' experiment.

REFERENCES

Happer, W., and Mathur, B. S. (1967*a*). Effective operator formalism in optical-pumping. *Phys. Rev.*, **163**, 12.

——— (1967*b*). Coherent detection of a new type of optical double-quantum transition. *Phys. Rev. Lett.*, **18**, 727.

Mathur, B. S., Tang, H., and Happer, W. (1968). Light shifts in the alkali atoms. *Phys. Rev.*, **171**, 11.

——— (1970). Light propagation in optically pumped alkali vapours. *Phys. Rev.*, A **2**, 648.