

change preclude extrapolation; enhancements σ_{100} are presented for the highest available rf power, 100 W ($H_{1e} = 0.7$ G). The shapes of the saturation curves as reflected in H and F signals were identical, so comparison of enhancements is valid.

The observed enhancements for H and F are seen to be nearly equal for both asphaltene and DPPH in $C_6H_5CF_3$. The difference in enhancements in DPPH- $C_6H_5CF_3$ solutions is partially due to the less favorable leakage factor for F nuclei; the bulk relaxation time of F is 2.7 sec; of H, 16 sec. On the other hand, the enhancements are quite different for H and F in the C_6F_6 - C_6H_6 samples. The difluorobenzene is an intermediate case. Relaxation both for H and F was dominated by the electron in all solutions. This indicates that both nuclei come within the same effective distance from the electron, and that repulsion of fluorine nuclei is not significant in these samples.

Substitution of deuterated benzene for normal benzene enables an estimate of the indirect transfer of electron polarization to fluorine via hydrogen. The fluorine enhancement by DPPH is the same in either deuterated or normal solutions. In asphaltene solutions, the net dipolar (negative) enhancement for fluorine in a mixture of $C_6D_6 + C_6F_6 + CS_2$ was slightly higher than in the corresponding hydrogenated sample, but nonetheless still very small. It may thus be concluded in the case of C_6F_6 that the feeble fluorine enhancements are not due

to indirect polarization via the protons, nor to an especially effective fluorine relaxation by protons.

The enhancement-saturation curve for F in the solutions of asphaltene with C_6F_6 was very curious. The F signal initially declined, as expected, but as the power was increased beyond a few watts, the signal commenced to grow again, finally reaching a small positive enhancement. A similar result was obtained in a corresponding deuterated sample (no. 7). The F signal from the DPPH + $C_6D_6 + C_6F_6$ sample behaved normally. The unusual saturation curve in the asphaltene- C_6F_6 samples is presumably due to a property of some of the assorted radicals present in asphalt.

While we can offer no detailed explanations at this time, the data suggest that the polarization of fluorine nuclei depends on their location in the solvent molecule, with a particular difference between aromatic and aliphatic positions. Aromatic F nuclei in solutions of DPPH indicate at least partial relaxation from a scalar ($A\vec{s}\cdot\vec{I}$) interaction.³ It would be expected that the methyl F nuclei should experience less scalar interaction than the ring nuclei, and this seems to be the situation here.

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SPIN-EXCHANGE LIGHT MODULATION BY ATOMIC HYDROGEN*

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It is well known that spin exchange may be used to create and detect polarization in an atomic system in the presence of an optically pumped vapor.^{1,2} However, it seems not generally understood that a transfer of coherence or "off-diagonal" information³ can be accomplished by the spin-exchange mechanism.

We have observed a transfer of coherence from atomic hydrogen, which is driven in magnetic resonance, to optically pumped sodium vapor. The effect appears as a modulation of sodium monitoring light at a frequency corre-

sponding to resonance in the hydrogen ground state. This experiment thus bears the same relation to the experiments of Anderson, Pipkin, and Baird, Jr.,² as do those of Dodd and Series,³ or Bell and Bloom,⁴ to the ordinary optical pumping experiments of Baratt, Brosel, and Kastler.⁵ It permits the direct observation of atomic-hydrogen precession without using Lyman alpha radiation.

A schematic of the apparatus is shown in Fig. 1. Sodium-pumping light is produced by an electrodeless rf discharge lamp, and the

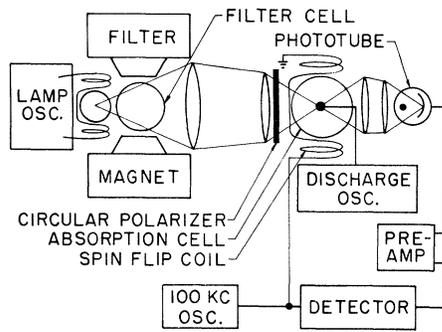


FIG. 1. A schematic of the apparatus. The static magnetic field is applied in the plane which includes the optic axis and is perpendicular to the page.

D_2 line is removed by a Zeeman-effect filter cell.⁶ After passing through a circular polarizer, the light is incident on an absorption cell containing sodium metal, about 2 Torr hydrogen, and 24 Torr helium buffer gas. Atomic hydrogen is produced by a weak steady rf discharge, and a several turn spin-flip coil wound around the absorption cell is driven at $\omega_0 = 100$ kc/sec to produce magnetic resonance in either sodium or hydrogen, depending on the value of the static magnetic field. The dc magnetic field is applied at about 30° to the optic axis by three mutually perpendicular sets of Helmholtz coils. Signals generated in a phototube by the transmitted light are amplified by a pre-amplifier tuned to ω_0 and fed into a phase-sensitive detector. The phase reference signal for the detector is taken from the oscillator which drives the spin-flip coil. As the main magnetic field is varied, light modulation is observed not only when the sodium is in resonance at 700 kc/sec/G, but also when atomic hydrogen is produced and in resonance at 1.4 Mc/sec/G.

In the limit in which the exchange collision between hydrogen and sodium occurs in a time short compared to the Larmor period of either system, clearly valid here, a simple theory of the exchange modulation and its line shape can be given. A density matrix is developed for the polarized atomic-hydrogen ground state in which the $F = 1, M_F = 1, 0, -1$ substates are driven in steady-state Larmor precession. At the time of the collision, t_0 , the electron is decoupled from the proton and exchanged with an electron from an unpolarized sodium atom. This is represented by using an exchange operator in the F, M_F representation, or more

simply by using Wigner vector-coupling coefficients. The steady-state density matrix of the sodium atom at time t is obtained by integrating the effect of spin exchange over the range $-\infty < t_0 < t$, and including a damping factor, $\exp[-\gamma(t-t_0)]$, where γ is the effective $1/T_2$ for sodium. In the final density matrix most, but not all, of the time dependence of the atomic-hydrogen resonance is preserved. The optical-exchange modulation signal for the case of a circularly polarized cross beam of the Bell-Bloom type is

$$S = (\text{const.})(\gamma^2 + \Delta^2)^{-1}[(\gamma \cdot D + \Delta \cdot A) \cos \omega_0 t + (\Delta \cdot D + \gamma \cdot A) \sin \omega_0 t],$$

where $\Delta = \omega_H - \omega_{Na}$ and D and A are dispersion and absorption "Bloch" functions of the hydrogen resonance. A simple theory of this type seems to account for the somewhat related observation that nuclear resonance in the ground state of He^3 produces optical modulation at the nuclear Larmor frequency in the light used to pump the metastable He^3 state.⁷ The theory and experiment emphasize that the spin-exchange collision, though acting as a relaxation process for the driven system, is itself a "driving" interaction, not only for the z component of polarization of an initially unpolarized system, but for the x and y spin components as well, a fact arising from the $\vec{S}_1 \cdot \vec{S}_2$ form of the interaction.

This experiment suggests that perhaps a similar technique can be used to observe level crossing effects⁸ and the excited-state structure of spectroscopically inaccessible atoms. This type of exchange modulation may also be useful in determining spin-exchange-induced line shifts and linewidths, the importance of which has been recently discussed by Bender.⁹

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INTERFERENCE OF FINE-STRUCTURE LEVELS IN HYDROGEN*

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We have observed interference between hydrogen fine-structure levels for $n=4, 5, 6,$ and 7 . The experiment was originally designed by Bickel to determine whether excitation equilibrium occurs when energetic ions pass through carbon foils. H_3^+ particles at 200 keV were sent through two successive foils; each foil was $\sim 10 \mu\text{g}/\text{cm}^2$ thick. Photographs were taken of the integrated light and of the individual Balmer lines, H_α through H_ϵ , which the foil-excited hydrogen atoms radiated. We then studied the spectrum from excited particles disturbed by electrostatic and magnetostatic fields. The magnetic field appeared between the foils as well as beyond the second one; the electric field, applied between two parallel metal plates which straddled the beam, did not extend between the foils. Experiments were done with H^+ , H_2^+ , and H_3^+ particles, and with single, double, and triple foils.

Figure 1(a) shows the integrated light from a beam which passed through two foils. The effect of the second foil is substantially different from that of the first. Hence, the foils do not produce excitation equilibrium. This negates Kay's assumption¹ that such equilibrium is achieved.

Figure 1(b) shows Balmer lines from a single-foil run. The spectrograph slit was wide open. The lines are slanted because of the Doppler effect.

Figure 1(c) shows the lines which resulted from sending the beam through two foils when an 8-G transverse magnetic field was applied. This field gave a motional transverse electric field of 29 V/cm. Figure 1(d) shows the effect of a transverse electric field of 51 V/cm.

Figures 1(c) and 1(d) show regularly spaced bright spots on the spectral lines. The clear-

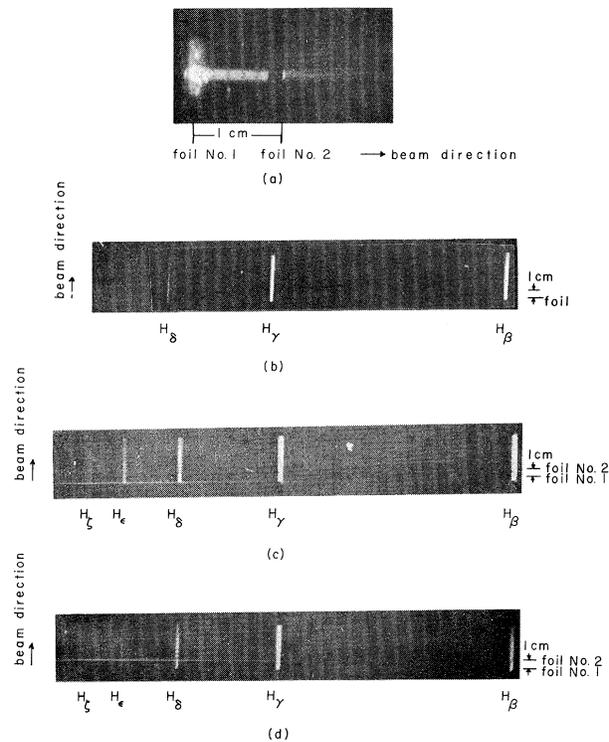


Fig. 1. (a) Photograph of integrated light from 200-keV H_3^+ particles incident on foil no. 1. The particles transmitted through foil no. 1 then pass through a second foil. The bright spot most clearly seen beyond the second foil is believed to be transition radiation. The integrated light exhibits a sharp intensity decrease after the second foil. (b) Resolved Balmer lines. The incident beam of 200-keV H_3^+ particles passed through a single foil. (c) Resolved Balmer lines when two foils were used with the 200-keV H_3^+ incident beam and an 8-G transverse magnetic field was present. Note the spots beyond the second foil. These spots are not present between the foils. (d) As in (c), with the magnetic field replaced by a transverse electric field of 51 V/cm.