POLARIZATION OF STABLE AND RADIOACTIVE NOBLE GAS NUCLEI BY SPIN EXCHANGE WITH LASER PUMPED ALKALI ATOMS

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POLARIZATION OF STABLE AND RADIOACTIVE NOBLE GAS NUCLEI BY SPIN EXCHANGE WITH LASER PUMPED ALKALI ATOMS

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Résumé - Les noyaux des gaz rares peuvent être fortement polarisés par échange de spin avec des vapeurs alcalines suffisamment denses et polarisées par pompage optique. Seule une faible fraction du moment angulaire de spin des atomes alcalins est transférée vers le spin nucléaire des atomes de gaz rare. La plus grande partie de ce moment disparait dans le mouvement relatif de l'atome alcalin par rapport à l'atome de gaz rare. Pour les gaz rares lourds, la plus grande partie du transfert de moment angulaire a lieu dans la molécule de Van der Waals formée par le couple alcalin-gaz rare. L'efficacité de ce transfert est déterminée par le taux de formation et de destruction des molécules de Van der Waals dans le gaz ambiant. Nous avons développé des méthodes expérimentales de mesure de l'efficacité du transfert du spin. Des noyaux de gaz rares radioactifs ont été polarisés par ces méthodes et la polarisation a été détectée grâce à l'anisotropie des produits de la désintégration radioactive. Nous en avons déduit des valeurs très précises des moments magnétiques des noyaux radioactifs.

Abstract - The nuclei of noble gases can be strongly polarized by spin exchange with sufficiently dense optically pumped alkali vapors. Only a small fraction of the spin angular momentum of the alkali atoms is transferred to the nulcear spin of the noble gas. Most of the spin angular momentum is lost to translational angular momentum of the alkali and noble gas atoms about each other. For heavy noble gases most of the angular momentum transfer occurs in alkali-noble-gas van der Waals molecules. The transfer efficiency depends on the formation and breakup rates of the van der Waals molecules in the ambient gas. Experimental methods to measure the spin transfer efficiencies have been developed. Nuclei of radioactive noble gases have been polarized by these methods, and the polarization has been detected by observing the anisotropy of the radioactive decay products. Very precise measurements of the magnetic moments of the radioactive nuclei have been made.

In 1960 Bouchiat Carver and Varnum[1] showed that it was possible to transfer spin angular momentum from optically pumped Rb vapor to the nuclei of ^{3}He buffer gas by collisions. Unfortunately, the transfer rates were very slow and many hours were required to build up detectable nuclear spin polarization in ^{3}He. Some 18 years later Grover[2] reported that the spin transfer rates from optically pumped Rb vapor to the nuclei of the heavy noble gases ^{129}Xe and ^{83}Kr were many orders of magnitude faster, and substantial nuclear spin polarization could be built up in a few minutes of optical pumping in favorable cases.

Once the noble gas nucleus is spin polarized it can retain its polarization for a very long time, hours or even days in cells with properly treated walls. Such slowly relaxing spins can be used to make very precise measurements of small terms in the spin Hamiltonian of the noble gas. For example, if the relaxation time is one hour and the signal to noise ratio is 100, one can measure the resonance frequency to a statistical uncertainty of $\pi^{-1} \times (1\ \mathrm{hr})^{-1} \times (100)^{-1} \simeq 1\mu$ Hz. Thus, one can easily sense the rotation rate, 11.6 Hz, of the earth, and serious efforts have been made to develop commercial gyroscopes based on spin polarized noble gas

nuclei.[3] Small upper bounds on the electric dipole moment of the ^{129}Xe nucleus have been set by looking for some influence of an external electric field on the nuclear magnetic resonce frequency.[4] There are other interesting uses of spin polarized noble gases in nuclear physics as targets for scattering experiments and as highly polarized sources of radioactive atoms. Interesting studies of surface interactions are possible.[5] It would probably be possible to liquify or freeze some of the heavy noble gases without much loss of polarization, and very interesting spin interactions could be expected in these highly polarized, condensed nuclear spin systems.

An important reason for the large enhancement of the spin transfer rates between optically pumped alkali atoms and heavy noble gases is the contribution of loosely bound alkali-noble-gas van der Waals molecules to the spin relaxation, a phenomenon first discovered by Aymar, Bouchiat and Brossel.[6] The nature of the interaction is indicated in Fig. 1

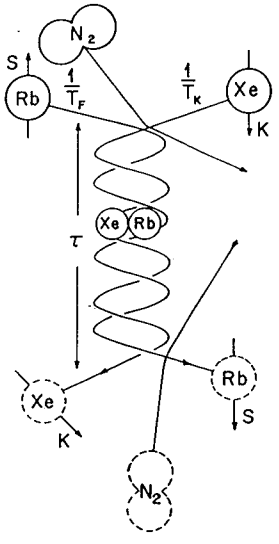


Fig. 1 - Alkali-noble gas van der Waals molecules are formed in three-body collisions at a rate of T_F^{-1} per alkali atom and T_K^{-1} per noble gas atom. They are broken up at a rate τ^{-1} by collisions with other atoms or molecules. Angular momentum can be transferred from the alkali electron spin S to the nuclear spin K of the noble gas atom during the relatively long molecular lifetime τ.

To investigate the physics of the spin transfer in van der Waals molecules we have made use of the apparatus[7] sketched in Fig. 2.

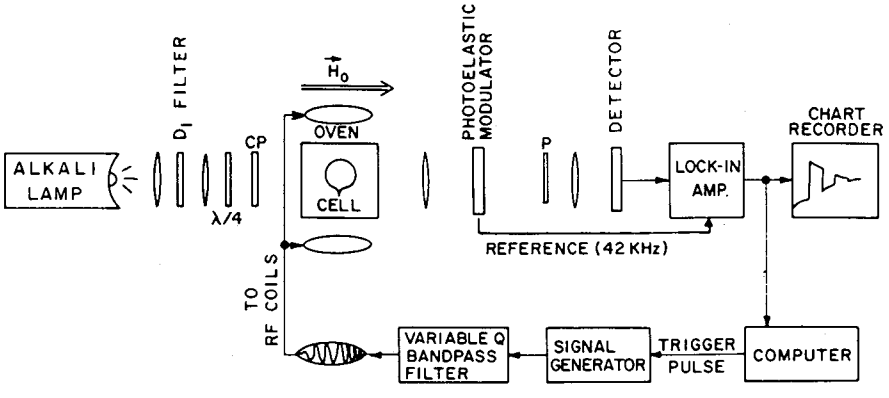


Fig. 2 - The apparatus used to study spin transfer between alkali atoms and noble gas molecules. CP is a circular polarizer; P is a linear polarizer used in conjunction with the photoelastic modulator to detect circularly polarized light; $\frac{\lambda}{4}$ is a compensator plate to null out stray circular polarization during the probe phase.

For systematic studies of spin transfer to ^{129}Xe we prefer not to use a laser, but instead to use a traditional, quiet and inexpensive electrodeless discharge lamp.[8] Laser pumping is essential for studies of other noble gas isotopes and especially for studies of radioactive isotopes. Each circularly polarized D_1 photon absorbed by the vapor deposits approximately $\frac{\hbar}{2}$ units of angular momentum in the alklai atoms. A certain fraction of this angular momentum is transferred to the nuclei of the noble gases, mainly during the lifetime of van der Waals molecules sketched in Fig. 1. The largest part of the angular momentum is lost to $\vec{N}$, the rotational angular momentum of an alkali-noble gas pair about each other. The spin transfer rates are so slow that we ordinarily wait for 10 or 15 minutes for equilibrium nuclear polarization to be established in the noble gases. Once the spins are polarized we remove the circular polarizer from the input optics of Fig. 2 so that unpolarized D_1 light enters the cell. During this probe phase, spin angular momentum flows back from the noble gas nuclei to the electron spins of the Rb atoms and a small amount of spin polarization is maintained in the alkali vapor. This alkali polarization in turn induces a weak circular polarization of the probe light which is detected with the photoelastic modulator. To eliminate the effects of slow drifts in the electronic amplifiers and in the thermally-induced polarizing properties of the optical elements we periodically invert the noble gas polarization with a chirpped audiofrequency pulse. Representative signals are shown in Fig. 3.

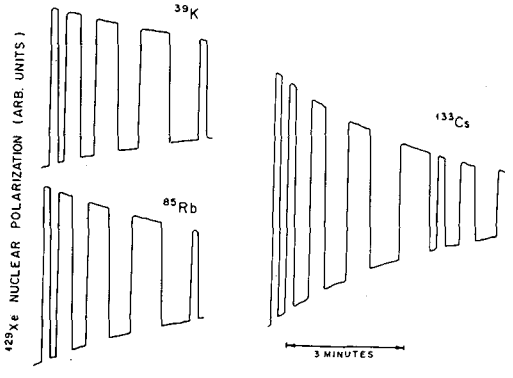


Fig. 3 - Decaying ^{129}Xe nuclear spin polarization observed in K, Rb and Cs vapor. The nuclear polarization is inverted periodically to eliminate the effect of slow drifts in the recording system. By using different intervals between inversions the slight spin destruction per inversion ($\lesssim 1\%$) can be accounted for.

Decay transients such as those of Fig. 3 can be studied as a function of the temperature of the sample cell. We find that the nuclear spin relaxation rate is a linear function of the alkali number density as inferred from saturated vapor pressure curves.[9] Representative data is shown in Fig. 4.

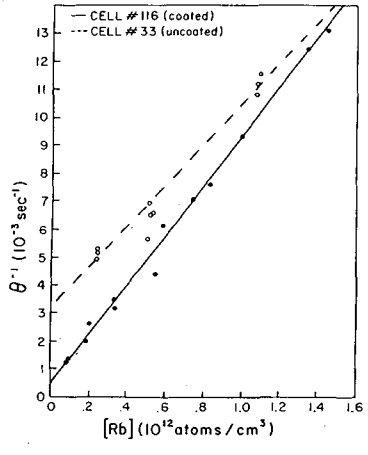


Fig. 4 - Spin relaxation of ^{129}Xe as a function of alkali number density. The wall-coated cell contained 14.9 Torr of N_2 and the uncoated cell contained 21 Torr of N_2. Both cells contained 0.5 Torr of Xe, isotopically enriched to 69% ^{129}Xe.

The extrapolated rate at zero alkali density is due to spin interactions on the cell walls. This wall relaxation can be significantly suppressed with silicone coatings.[10]

The measured relaxation rates depend on the third-body pressure as one would expect from the physical picture sketched in Fig. 1. At low third-body pressures the rates are slow because the molecules form too slowly. At high third-body pressures the rates are again slow because the molecules are broken up too quickly. An example of the dependence of the rates on third body pressure is shown in Fig. 5.

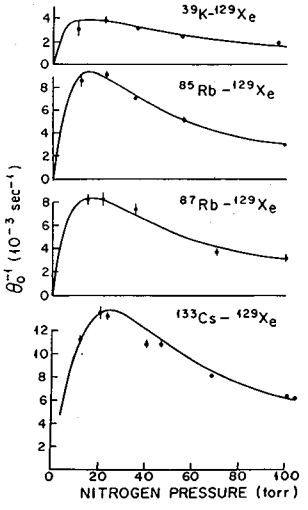


Fig. 5 - Measured spin relaxation rates of ^{129}Xe in various alkali vapors as a function of nitrogen pressure. All rates are given for an alkali number density of 10^{12} atoms/cm^2 as inferred from saturated vapor pressure curves.[9]

The relaxation rate also depends strongly on the magnitude of the external magnetic field in which the sample cell is located. An example is shown in Fig. 6.

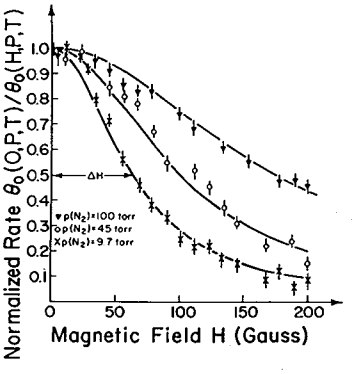


Fig. 6 - Slowing down of the relaxation rate of ^{129}Xe in ^{87}Rb vapor in an external magnetic field H. The rates are normalized to their low-field values.

The slowing down of the nuclear spin relaxation occurs for the same reason as the slowing down of the electronic spin relaxation of Rb in various buffer gases first observed and explained by Bouchiat et al.[11] The magnetic field decouples the electronic spin $\vec{S}$ of the Rb atom from the rotational angular momentum $\vec{N}$ in the long-lived van der Waals molecules.

The simplest spin Hamiltonian which is consistent with presently known data on spin relaxation and spin transfer in mixtures of alkali vapors and noble gases is

$$H = A\vec{I}\cdot\vec{S} + \gamma\,\vec{N}\cdot\vec{S} + \alpha\,\vec{K}\cdot\vec{S} + g_S\mu_B\vec{B}\cdot\vec{S} + g_I\mu_B\vec{B}\cdot\vec{I} + g_K\mu_B\vec{B}\cdot\vec{K} + \ldots\ldots \qquad (1)$$

The physical significance of the various angular momentum vectors of (1) is illustrated in Fig. 7.

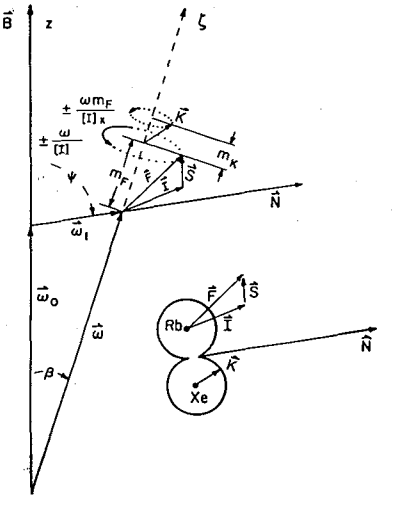


Fig. 7 - A vector model of the spin coupling of Eq. 1. The spin rotation frequency is $\omega_1 = \gamma N/h$ and the electron Larmer frequency in the external field is ω_0. The Breit-Rabi parameter of Eq. 1 is $x = \frac{\gamma N}{\alpha}$. The statistical weight of the alkali nucleus is $[I] = 2I+1$.

The electronic spin $\vec{S}$ of the alkali atom in the molecule is coupled to the nuclear spin $\vec{I}$ of the alkali atom by the magnetic dipole interaciton $A\vec{I}\cdot\vec{S}$. The electron spin of the alkali is also coupled to the rotational angular momentum $\vec{N}$ of the molecule by spin-rotaton interaction $\gamma\vec{N}\cdot\vec{S}$. The nuclear spin of K of the noble gas atom is coupled to the electron spin of the alkali by the magnetic dipole interaction $\alpha\vec{K}\cdot\vec{S}$. An external magnetic field B couples to the magnetic moments of S, I and K as shown in (1). The g factors g_I and g_K will be some three orders of magnitude smaller than g_S. All of the coupling coefficients, A, γ, a..., will depend somewhat on the vibrational and rotaitonal state of the van der Waals molecule or on the internuclear separation and velocity of an unbound colliding pair.

By systematically investigating the spin relaxation in various alkali noble gas pairs with the methods outlined above we have been able to determine the key parameters which govern the spin transfer rates in a number of alkali-noble gas systems. For example, for $^{87}Rb/^{129}Xe$ we find

Breit-Rabi parameter: $x = \frac{\gamma N}{\alpha} = 3.1(3)$

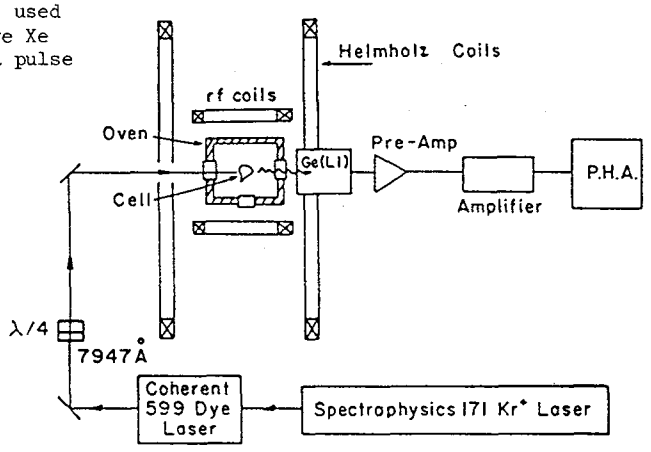
Pressure-lifetime product: $\tau p(N_2) = 1.5(2)\times10^{-7}$ sec Torr

3-Body formation rate: $\frac{1}{T_K[Rb][N_2]} = 4.8(5)\times10^{-32}cm^6sec^{-1}$

Spin-rotation coupling: $\frac{\gamma N}{h} = 110(10)$ MHz

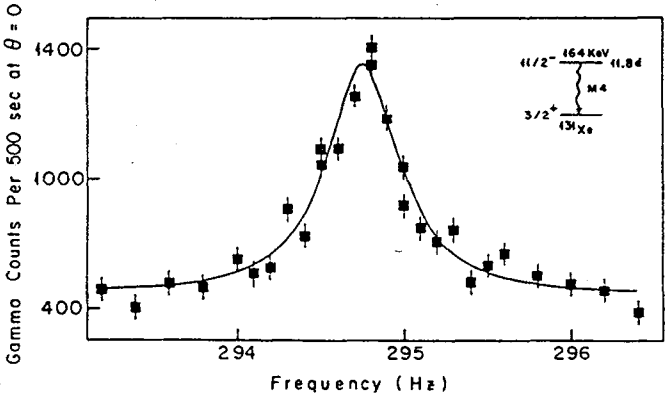
We have recently extended these polarization methods to radioactive Xe isotopes. Our apparatus is shown in Fig. 8.

Fig. 8 - The apparatus used to polarize radioactive Xe isotopes. P.H.A. is a pulse height amplifier.



Pyrex sample cells, 12 mm in diameter were prepared by distilling a small amount of ^{87}Rb metal into the cells and adding about 10 μ Ci of radioactive ^{133}Xe along with traces of ^{131m}Xe and ^{133m}Xe. The isotopes were purchased from the General Electric Company in glass ampules. About 50 Torr of N_2 gas was also added to the cell to serve as a third body and to quench the fluorescence of laser excited atoms. The cell was heated to a temperature of 150°C to provide adequate spin exchange rates. At these temperatures the cell is some 100 optical depths thick so laser optical pumping is essential to burn through the vapor and maintain high alkali spin polarization. A lithium-drifted germanium detector was used to detect gamma rays, and when the Xe nuclei were spin polarized the spatial distribution of the gamma rays changed. Thus we could use the changes in the anisotropic gamma radiation to detect nuclear magnetic resonance in the radioactive Xe atoms. An example of such a reesonance is shown in Fig. 9.

Fig. 9 - Nuclear resonance of ^{131m}Xe observed with the apparatus of Fig. 8.



The excellent signal to noise ratio of Fig. 9 shows that spin exchange laser pumping of radioactive noble gas nuclei is a promising experimental tool which will be useful in many other experiments.

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