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Spin exchange optical pumping of hydrogen and deuterium nuclei

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Abstract

The prospects for the direct production of a nuclear polarized atomic hydrogen or deuterium target by the use of spin exchange optical pumping is analyzed in both a low and a high magnetic field. In a low magnetic field it is found that the optical pumping cell wall coatings must have relaxation times that correspond to thousands of wall collisions if one is to produce a highly polarized target. In a high magnetic field it is found that wall coatings must have relaxation times that correspond to only a few hundred wall collisions in order to produce a highly polarized atomic hydrogen target but must have relaxation times that correspond to thousands of wall collisions to produce a highly polarized atomic deuterium target.

1. Introduction

There is currently high interest in the production of a nuclear spin polarized atomic hydrogen or deuterium target for use as an internal target in storage rings. One promising method for producing a nuclear spin polarized target is by the use of spin exchange optical pumping. Coulter et al. [1] have obtained a flux of 2.1×10^{17} deuterium atoms/s with an electron spin polarization of $73 \pm 3\%$ by the use of spin exchange optical pumping. In a previous paper we analyzed the effect of radiation trapping on a high field spin exchange optically pumped atomic hydrogen or deuterium target [2]. In that paper it was assumed that the magnetic field was high enough that the electron and nuclear spins were completely decoupled so that hydrogen–hydrogen (or deuterium–deuterium) spin exchange collisions do not result in the polarization of the nuclear spins. Since that time we have studied the consequences of spin exchange collisions between the hydrogen atoms in a target at magnetic fields where the electronic and nuclear spins are not completely decoupled so that the nuclear spins become polarized [3,4]. In this paper we analyze the constraints on a spin exchange optically pumped nuclear spin polarized target operated in a magnetic field that does not completely decouple the electronic and nuclear spins so that spin exchange collisions result in the transfer of electronic polarization into nuclear polarization. We consider limitations for a spin exchange optically pumped target in both a low magnetic field and in a high but not infinitely large magnetic field so that the electronic and nuclear spins are not completely decoupled. The results in this paper are identical to those of Ref. [2] in the limit of infinite magnetic field.

A nuclear polarized H target can be made as follows. Molecular hydrogen is partially dissociated and flows into an optically pumped cell that contains an alkali vapor. The alkali vapor and atomic hydrogen flow through the optical pumping cell where the alkali atoms are polarized by optical pumping. The atomic hydrogen is polarized by spin exchange collisions with the alkali atoms (A–H spin exchange collisions). In the cell there are also hydrogen–hydrogen (H–H) spin exchange collisions and alkali–alkali (A–A) spin exchange collisions. The hydrogen atoms can lose their polarization by collisions with the cell walls, by collisions with other atoms or molecules, by recombination, or by flowing out of the cell. For a useful target the H atoms must lose their spin polarization primarily by flowing out of the optical pumping cell. The alkali atoms can also lose their spin polarization in a variety of ways, but for a useful target the alkali atoms must lose their polarization primarily through spin exchange collisions with hydrogen rather than by collisions with the walls, by collisions with other atoms or molecules, or by flowing out of the cell. Radiation trapping affects the optical pumping of the alkali and thereby affects the rate of polarization of the atomic hydrogen. Atomic deuterium can also be polarized by spin exchange optical pumping. After the atomic hydrogen is polarized it flows out of the optical pumping cell and into a storage cell target. This paper analyzes the various fundamental limitations on the production of a nuclear polarized atomic hydrogen or deuterium target by spin exchange optical pumping.

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In this paper we show that a nuclear spin polarized hydrogen or deuterium target can be obtained if the following conditions are met:

(i) The electronic spin relaxation time for the alkali atoms is long compared to the A–H spin exchange time.

(ii) The electronic spin relaxation time for the hydrogen atoms is long compared to the average dwell time of a hydrogen atom in the optical pumping cell.

(iii) The A–H spin exchange time is short compared to the average dwell time of a hydrogen atom in the optical pumping cell.

(iv) The time for the hydrogen atoms to approach spin temperature equilibrium is short compared to the average dwell time for a hydrogen atom in the optical pumping cell. This condition is strongly dependent on the magnetic field at the optical pumping cell since the time for the hydrogen atoms to approach spin temperature equilibrium is directly proportional to $1+x^2$ where x is the ratio of the magnetic field to the critical field for decoupling of the electronic and nuclear spins of the hydrogen atom in the ground level. For hydrogen and deuterium the critical fields are 507 and 117 G respectively.

(v) The flow rate of polarized hydrogen atoms in atoms/s from the optical pumping cell must be less than the number of photons/s absorbed by the alkali vapor divided by the average number of photons required to polarize an alkali atom.

(vi) In a low magnetic field the alkali density must be low enough that radiation trapping does not depolarize the alkali atoms. This results in the necessity for long average dwell times for the hydrogen atoms in the optical pumping cell and therefore very long hydrogen electronic spin relaxation times. This requires wall coatings that cause extremely small depolarization rates for the electronic spins of the hydrogen atoms per wall collision.

(vii) In a high magnetic field radiation trapping changes the number of photons required to polarize an alkali atom so that the rate of production of polarized hydrogen atoms can not exceed that allowed by the alkali density, and therefore the number of laser photons must be sufficient for the alkali density.

A preliminary version of some of the material in this paper was presented at the Workshop on Polarized Ion Sources and Polarized Gas Targets held in 1993 at Madison, Wisconsin [5].

2. The rate equations for spin exchange optical pumping

We discuss spin exchange optical pumping with circularly polarized alkali resonance radiation. The rate equations for the alkali electron spin polarization (P_A^e), the hydrogen electron spin polarization (P_H^e), and the hydrogen nuclear spin polarization (P_H^n), can be obtained by combining material in Ref. [2] and Ref. [4]. The rate equations are the following:

$$\frac{dP_A^e}{dt} = \frac{R}{N}(1-P_A^e) - \frac{n_H}{n_A}\frac{1}{T_{AH}}(P_A^e - P_H^e), \tag{1}$$

$$\frac{dP_h^e}{dt} = \frac{1}{T_{AH}}(P_A^e - P_H^e) - \frac{1}{2T_{ST}}(P_H^e - P_H^n) - \frac{P_H^e}{T_H}, \tag{2}$$

and

$$\frac{dP_H^n}{dt} = \frac{1}{2T_{ST}}(P_H^e - P_H^n) - \frac{P_H^n}{T_H}, \tag{3}$$

where n_A and n_H are the alkali and atomic hydrogen number densities respectively, R is the laser absorption rate per atom in photons/s, N is the average number of photons needed to polarize an alkali atom, T_H^{-1} is the total hydrogen polarization loss rate in the target, T_{AH}^{-1} is the A–H spin exchange rate; and T_{ST} is the H–H spin temperature equilibrium rate. The total hydrogen polarization loss rate in the target is made up of two terms $T_H^{-1} = T_{HR}^{-1} + T_{HD}^{-1}$ where T_{HR}^{-1} is the spin relaxation rate and T_{HD}^{-1} is the loss rate due to flow out of the optical pumping cell. In order to obtain optimum operation of the spin exchange polarized target the hydrogen loss rate must be dominated by the flow out of the optical pumping cell so that $T_H^{-1} \approx T_{HD}^{-1}$. Thus for optimum operation of the target T_H should be nearly equal to the average dwell time for a hydrogen atom in the optical pumping cell, T_{HD}. The A–H spin exchange rate is given by

$$\frac{1}{T_{AH}} = n_A \langle \sigma_{SE} v \rangle_{AH}, \tag{4}$$

where $\langle \sigma_{SE} v \rangle_{AH}$ is the average of the A–H spin exchange cross section times the relative A–H velocity over the velocity distribution function for the A–H collisions. The H–H spin temperature equilibrium rate is given by

$$\frac{1}{T_{ST}} = \frac{n_H \langle \sigma_{SE} v \rangle_{HH}}{1+x^2}, \tag{5}$$

where $\langle \sigma_{SE} v \rangle_{HH}$ is the average of the H–H spin exchange cross sections times the relative velocity over the velocity distribution function for the H–H collisions. Note that T_{ST} depends on x and hence on the magnetic field. Eqs. (1)–(3) are strictly correct only in a high magnetic field, but they offer a reasonable approximation for P_H^e and P_H^n in a low magnetic field.

In writing the rate equations we have ignored the alkali nuclear spin. In the limit of rapid optical pumping (i.e. when the alkali pumping rate is large compared to the A–H spin exchange rate) ignoring the nuclear spin is reasonable since the alkali nucleus is always highly polarized and the alkali electron spin polarization is lost to the H atoms via spin exchange collisions and then rapidly replaced by the optical pumping. We have also ignored alkali depolarization by collisions with the cell walls, by

collisions with other atoms or molecules, or by flowing out of the cell. For a target to be useful it is essential that the angular momentum of the optical pumping light be passed rapidly through the alkali vapor into the hydrogen. It is essential that $n_H/(n_A T_{AH})$ be much larger than all other polarization loss rates from the alkali. We again stress that the main hydrogen polarization loss mechanism should be flow out of the optical pumping cell. If the hydrogen in the polarized target downstream from the optical pumping cell is to have a high density and polarization it is essential that the primary hydrogen polarization loss rate is due to flow out of the cell rather than relaxation.

3. Low magnetic fields

In the experiment of Coulter et al. a high magnetic field (2.2 kG) is used at the optical pumping cell [1]. The high magnetic field permits the optical pumping to be carried out at relatively high alkali densities whereas radiation trapping would cause severe depolarization in a low magnetic field. Radiation trapping acts to depolarize an optically pumped alkali vapor in a low magnetic field so that one can not obtain a high polarization in a low magnetic field at high alkali density. In contrast to its depolarizing effect in a low magnetic field, radiation trapping does not act to depolarize an optically pumped alkali vapor in a high magnetic field. Thus in a high magnetic field an alkali vapor can be polarized even in the presence of radiation trapping. The effect of radiation trapping in a high magnetic field is to require more photons per atom to polarize the alkali vapor than are required at an alkali density low enough that radiation trapping is not significant [6,7].

One might ask whether it is possible to construct a nuclear polarized hydrogen or deuterium target with the spin exchange optical pumping in a low magnetic field. We analyze this question in this section. In order to consider the problem let us assume that the optical pumping cell is a right circular cylinder of radius r and length L where r is less than L. In order to avoid severe radiation trapping it is necessary that the alkali density be such that $N_A \leq 1/(\sigma_{ABS} r)$ where σ_{ABS} is the alkali resonance absorption cross section at line center. In order to make the discussion concrete we consider the optical pumping of K. The $4^2S_{1/2} \rightarrow 4^2P_{1/2}$ absorption transition has a wave length of $\lambda = 770$ nm and an Einstein coefficient of $A = 3.82 \times 10^7\ s^{-1}$. The optical absorption cross section for a transition is given by

$$\sigma_{ABS} = \frac{g_u}{g_l}\frac{\lambda^2}{8\pi} A g(0), \tag{6}$$

where g_u and g_l are the statistical weights of the upper and lower levels respectively and $g(0)$ is the normalized line shape function evaluated at line center. For the $4^2P_{1/2} \rightarrow 4^2S_{1/2}$ transition $g_u = g_l = 2$. The transition has a Doppler broadened line shape. For a Doppler line shape $g(0) = 0.94/\Delta\nu_D$ where $\Delta\nu_D$ is the full width at half maximum of the Doppler line shape. The Doppler line width at a temperature of $T = 400$ K is $\Delta\nu_D = 8.3 \times 10^8$ Hz. Thus for the K resonance transition $\sigma_{ABS} = 9.5 \times 10^{-12}\ cm^2$.

The maximum spin exchange rate between the alkali vapor and the H atoms in a low magnetic field is

$$\frac{1}{T_{AH}} = n_A \langle \sigma_{SE} v \rangle_{AH} \approx \frac{\langle \sigma_{SE} v \rangle_{AH}}{\sigma_{ABS} r}. \tag{7}$$

For K at $T = 400$ K the value of $\langle \sigma_{SE} v \rangle_{AH}$ is $2.3 \times 10^{-9}\ cm^3/s$ and $\langle \sigma_{SE} v \rangle_{AH}$ changes only slowly with the temperature. If one takes $r = 1$ cm then the maximum alkali-hydrogen spin exchange rate is $1/T_{AH} = 2.5 \times 10^2\ s^{-1}$.

In order for the hydrogen atoms to become highly polarized it is obviously necessary that $1/T_{AH} \gg 1/T_H$, or using Eq. (7) one obtains

$$T_H \gg \frac{\sigma_{ABS} r}{\langle \sigma_{SE} v \rangle_{AH}}. \tag{8}$$

For a long cylinder the average wall collision rate for a hydrogen atom is equal to $v_H/2r$ where v_H is the mean H atom velocity. At $T = 400$ K and for $r = 1$ cm the wall collision rate for a hydrogen atom is about 1.5×10^5 wall collision/s. If the average number of collisions a hydrogen atom makes with the wall during a dwell time is M then $T_H \approx T_{HD} = (2rM)/v_H \gg (\sigma_{ABS} r)/\langle \sigma_{SE} v \rangle_{AH}$. The relative velocity v_{AH} is nearly the same as the atomic hydrogen velocity, v_H, since a potassium atom is about 39 times as massive as a hydrogen atom. If we take $\langle \sigma_{SE} v \rangle_{AH} = \sigma_{SE} v_{AH} \approx \sigma_{SE} v_H$ then we find that $M \gg \sigma_{ABS}/2\sigma_{SE}$. The H–K spin exchange cross section at thermal velocities is about $1.2 \times 10^{-14}\ cm^2$ so that $M \gg 7.8 \times 10^2$.

We can make this discussion more quantitative by considering the steady state solution to Eqs. (2) and (3). Although Eqs. (2) and (3) are correct only in a high magnetic field and the low magnetic field equations are somewhat more complicated than Eqs. (2) and (3) a reasonable estimate for the ratio P_H^e/P_A^e can be obtained using Eqs. (2) and (3). In the steady state where $dP_H^e/dt = dP_H^n/dt = 0$ these equations yield

$$\frac{1}{T_{AH}}(P_A^e - P_H^e) - \frac{P_H^e}{T_H} - \frac{P_H^n}{T_H} = 0. \tag{9}$$

In a low magnetic field T_{ST}^{-1} is approximately equal to $n_H \langle \sigma_{SE} v \rangle_{HH}$. For H–H collisions at $T = 400$ K the value of $\langle \sigma_{SE} v \rangle_{HH}$ is $4 \times 10^{-10}\ cm^3/s$ and the value is nearly independent of the temperature. For an atomic H number density of $n_H = 5 \times 10^{12}$ atoms/cm^3 the value of T_{ST} is 5×10^{-4} s. Thus $T_{ST} \ll T_H$. In this situation $P_H^e \approx P_H^n$ as can be seen from the steady state solution to Eq. (3). Substituting $P_H^e = P_H^n$ into Eq. (9) yields $P_H^e/P_A^e = (1 + 2T_{AH}/T_H)^{-1}$. This shows how P_H^e/P_A^e depends on (T_{AH}/T_H). If one requires that $P_H^e/P_A^e = 0.9$ then T_{AH}/T_H

$= 0.05$. For $T_{AH}/T_H = 0.05$ and for $T_{AH} = 4$ ms, which corresponds to the maximum spin exchange rate in a low magnetic field one finds that $T_H \approx T_{HD} = 80$ ms. A dwell time of 80 ms corresponds to about 10^4 wall collisions for the assumed size of the optical pumping cell ($r = 1$ cm and $r \ll L$). Thus it is possible to produce a spin exchange optically pumped nuclear polarized H target in a low magnetic field only if one can find a wall coating for the optical pumping cell that will allow the atomic hydrogen to make more than 10^4 wall collisions before spin relaxation occurs. It is clear that if the dwell time must be at least 80 ms then the hydrogen atom electronic spin relaxation time must be significantly greater than 80 ms. At the present there are no known wall coatings, that can survive interacting with K vapor and can permit more than 10^4 wall collisions before relaxation occurs [8]. Thus it seems that at the present it is necessary to use a high magnetic field at the optical pumping cell.

4. High magnetic fields

In a high magnetic field the spin temperature equilibrium rate, T_{ST}^{-1}, is approximately given by $(n_H \langle \sigma_{SE} v \rangle_{H-H})/x^2$. The approach to spin temperature equilibrium is slower than in a low magnetic field by the factor $1/x^2$. In this section we analyze the conditions under which it is possible to produce a nuclear spin polarized H target. Let us consider the steady state solution to Eqs. (1)–(3) in a high magnetic field. In the steady state where

$$\frac{dP_A^e}{dt} = \frac{dP_H^e}{dt} = \frac{dP_H^n}{dt} = 0 \tag{10}$$

the Eqs. (1), (2), and (3) can be solved. The solutions for P_H^e and P_H^n are the following

$$P_H^e = \frac{1}{1 + 2\left(\frac{1 + T_{ST}/T_H}{1 + 2T_{ST}/T_H}\right)\left(\frac{T_{AH}}{T_H} + \frac{n_H}{n_A T_H}\frac{N}{R}\right)}, \tag{11}$$

and

$$P_H^n = P_H^e \frac{1}{1 + \frac{2T_{ST}}{T_H}}. \tag{12}$$

It should be noted that the term $(1 + T_{ST}/T_H)/(1 + 2T_{ST}/T_H)$ varies only from $\frac{1}{2}$ when $T_{ST}/T_H = \infty$ to 1 when T_{ST}/T_H is 0. The electron spin polarization of the H atoms by spin exchange optical pumping in the limit where T_{ST}/T_H is very large has been treated by Anderson and Walker [3]. They assumed no nuclear spin polarization which is equivalent to assuming T_{ST}/T_H is infinite. In this paper we consider the prospects for producing directly nuclear spin polarization of the H atoms. This requires that $T_{ST} \ll T_H \approx T_{HD}$ as will be shown. In this situation $(1 + T_{ST}/T_H)/(1 + 2T_{ST}/T_H) \approx 1$. In order to have a high P_H^e it is necessary that $2T_{AH} \ll T_H$ and $2n_H N/n_A T_H R \ll 1$.

The first condition, $2T_{AH} \ll T_H \approx T_{HD}$, indicates that $n_A \langle \sigma v \rangle_{AH} T_H/2 \approx n_A \langle \sigma v \rangle_{AH} T_{HD}/2 \gg 1$ so that the alkali density is high enough that a hydrogen atom makes a sufficient number of spin exchange collisions with polarized alkali atoms during a dwell time in the optical pumping cell to acquire a polarization close to that of the alkali atoms. The value for $\langle \sigma v \rangle_{AH}$ for K and H spin exchange collisions is 2.3×10^{-9} cm^3/s at 400 K and is nearly independent of the temperature. Thus it is necessary that $n_A T_H \gg 9 \times 10^8$ s/cm^3. If we take n_A to be the same as for the Argonne target so that $n_A = 2 \times 10^{12}$ cm^{-3} then $T_H \gg 4.5 \times 10^{-4}$ s. If one desires P_H^e to be 0.9 or greater then it is necessary that $T_H \approx T_{HD} \geq 4.5$ ms provided $2n_H N/(n_A T_H R) = 0$. If $2n_H N/(n_A T_H R)$ is not zero then T_H must be even longer than 4.5 ms. This means that the dwell time must be at least 4.5 ms. A dwell time of 4.5 ms corresponds to about 400 wall collisions without depolarization for the assumed size of the optical pumping cell.

The second condition

$$2n_H N/(n_A T_H R) \ll 1 \tag{13}$$

leads to

$$F \ll \frac{P_L}{(2h\nu N)}, \tag{14}$$

where F is the flow rate of hydrogen atoms out of the optical pumping cell and P_L is the laser power. Eq. (14), the limitation on the flow rate, follows from Eq. (13), from the definition of the flow rate, $F = n_H V/T_{HD} \approx n_H V/T_H$, and from $R \leq P_L/(h\nu n_A V)$ where V is the volume of the optical pumping cell. Eq. (14) simply says that the flow rate of polarized H atoms from the optical pumping cell must be less than the flow rate of photons divided by the average number of photons needed to polarize an alkali atom.

If one obtains a large P_H^e then it is possible to have a large P_H^n provided $2T_{ST}/T_H \ll 1$ which is equivalent to $2x^2/(n_H \langle \sigma_{SE} v \rangle_{HH} T_H) \approx 2x^2/(n_H \langle \sigma_{SE} v \rangle_{HH}\ T_{HD}) \ll 1$. This condition also places a limitation on the flow rate out of the cell. Substituting the result that $T_{HD} = (n_H V)/F$ yields

$$F \ll \frac{n_H^2 \langle \sigma_{SE} v \rangle_{HH} V}{2x^2}. \tag{15}$$

This limitation indicates that the flow rate must be low enough that there are a sufficient number of H–H spin exchange collisions to polarize the nuclear spin. The spin exchange rate depends on the magnetic field through x. At very large fields the flow rate must be very slow. In order that the magnetic field is small enough for the nuclear polarization to be large it is necessary that

$$x \ll \sqrt{\frac{n_H^2 \langle \sigma_{SE} v \rangle_{HH} V}{2F}}. \tag{16}$$

The value of $\langle \sigma_{SE} v \rangle_{HH}$ at 400 K is 4×10^{-10} cm^3/s and

is relatively independent of the temperature. If we take $n_H = 10^{14}$ atoms/cm^3, $F = 2 \times 10^{17}$ atoms/s, and $V = 50$ cm^3 then we find that $x \ll 22$ or $B \ll 22 B_c = 11$ kG. In order that $P_H^n = 0.9\, P_H^e$ it is necessary that $B \leq 3.5$ kG for the assumed values of n_H, F, and V.

The first condition on the flow rate, $F \ll P_L/(2h\nu N)$, also depends on the magnetic field through the fact that the average number of photons needed to polarize an alkali atom depends on both the alkali density and the magnetic field since the radiation trapping depends on the magnetic field [4,5]. Coulter et al. have shown that it is possible to optically pump to a high polarization a K vapor with $n_A = 10^{12}$ atoms/cm^3 in the presence of atomic hydrogen with $n_H = 10^{14}$ atoms/cm^3 and for a flow rate of 2.1×10^{17} if the magnetic field is about 2 kG [1]. This high an alkali density can not be optically pumped to a high polarization in a low magnetic field. Since the various numerical values for n_A, n_H, T_D and V chosen for our calculations are similar to the target of Coulter et al. it seems clear that one can construct a nuclear polarized H target provided the magnetic field at the target is in the range 2 to 3.5 kG.

For deuterium (D) nuclei the small critical field changes the situation significantly. The ratio of the rate of polarization of D to H is approximately $[x_H^2 \langle \sigma_{SE} v \rangle_{HH} n_H]/[x_D^2 \langle \sigma_{SE} v \rangle_{DD} n_D] \approx 1/30$ under the same conditions. Thus higher densities or lower magnetic fields are necessary in order to achieve high polarizations of D, than are necessary for high polarization of H. In lower fields the optical pumping rate will be adversely affected by radiation trapping, thus limiting the flow. At higher densities (at constant flow) longer dwell times are necessary, so that relaxation processes will be more important. We note that in the Argonne optical pumping cell the D atoms make hundreds of wall collisions. For longer dwell times more wall collisions will occur and relaxation by wall collisions may be important. In addition other relaxation processes may begin to become important as well. For D if one satisfies the condition to obtain nuclear polarization the ground level Zeeman-hyperfine level populations will be those predicted by the spin temperature [2]. In this situation one will have both vector and tensor polarization. The tensor polarization will, however, be relatively small unless the spin temperature is very low.

5. Conclusions

The analysis presented in this paper indicates that it may be possible to produce directly a nuclear spin polarized atomic H or D target by spin exchange optical pumping in either a low or a high magnetic field. If the optical pumping is to be carried out in a low magnetic field then wall coatings that have relaxation times corresponding to thousands of wall collisions are necessary. If the optical pumping is to be carried out in a high field then it appears that atomic hydrogen can be polarized with wall coating that have relaxation times corresponding to a few hundred wall collisions for flow rates of a few times 10^{17} atoms/s provided the magnetic field at the optical pumping cell is between 2 and 3.5 kG. It is more difficult to produce directly a nuclear polarized D target than a nuclear polarized H target by spin exchange optical pumping and better wall coatings will be required for a D target than for a H target. For D it may be preferable to use a very high magnetic field at the optical pumping cell followed by rf transitions to produce a nuclear polarized D target. However, care must be taken if this scheme is used, since Walker and Anderson have pointed out that if spin exchange collisions occur in the low or intermediate magnetic field required for the rf transitions then spin temperature equilibrium will be approached [2], and this must be considered in estimating the nuclear polarizations that can be obtained.

Acknowledgements

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