COLLISION PROCESSES IN QUANTUM GASES

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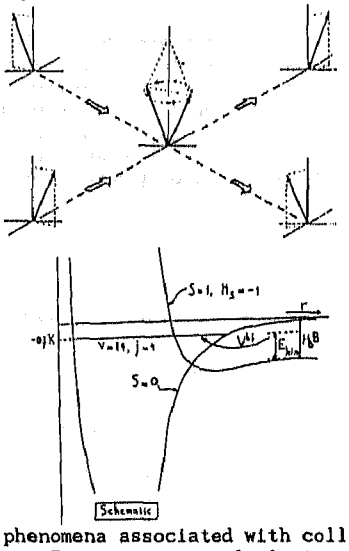
We discuss three topics relating to spin-polarized quantum gases: A) The role of the hyperfine interaction during collisions. B) The decay of doubly polarized atomic hydrogen (H↑↑) or deuterium (D↑↑) in a magnetic trap of arbitrary shape due to magnetic dipolar interactions taking into account the influence of degeneracy. C) The mechanism of three-body dipolar recombination $H + H + H \rightarrow H_2 + H$ in H↓↓.

1. Role of hyperfine interaction in collisions

Two- and three-body collisions play a central role in spin-polarized quantum gases. They are responsible for such diverse phenomena as spin waves, the spin-exchange frequency shift in the cryogenic H maser, two-body relaxation and three-body recombination. Spin-polarized H also offers one of the purest opportunities to study two-dimensional collisions with all the fascinating features that this implies in the quantum regime ($\lambda_{\text{de Broglie}}$) a): two-dimensional effective-range theory[1], formulated for the first time in view of the application to H gas adsorbed on a surface of superfluid helium. In this contribution this is the last word I have said on 2D H gas.

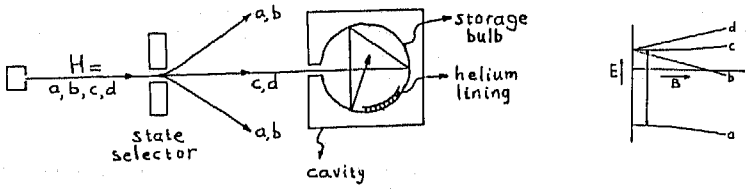
Already on the purely microscopic level a seemingly trivial problem like the collision of ground state H atoms presents unexpected challenge. This is due to the complicated dynamics of the four spins involved and to the subtle interplay between the extremely strong central exchange interaction and the very weak hyperfine interaction, each of which likes its own quantum numbers. On the basis of its extreme weakness one would expect the hyperfine interaction to be totally negligible during collisions. This is indeed so for a number of cases. For instance, to understand the essential physics of spin waves in atomic hydrogen, i.e. the identical spin rotation effect[2] of the proton spins, it is well-known that one can forget all dependence of the interaction on the proton spins and in particular on the hyperfine coupling.



On the other hand, the inverse predissociation effect in resonance recombination of H↓ is an example where the hyperfine interaction does play an essential role during collisions. The experiment[3] which showed for the first time that this effect plays a central role in the temperature dependence of two-body a + a recombination is another beautiful example of the rich variety of phenomena associated with collisions between such simple particles as H atoms. From accurate calculations on the basis of the most recent

Kolos-Wolniewicz potential including non-adiabaticity and relativistic and radiative corrections it appears that the (v,j) = (14,4) resonance through which the recombination is enhanced is most probably slightly unbound. If experiment would indicate more definitively a bound state (Cf. also the experiment on the B-dependence[4]), this might point to some fundamental effect which is still missing in the theoretical description.

Forgetting about the role of the hyperfine coupling has turned out to be an essential omission also in the case of the cryogenic H maser[5]. Let me make clear why this is the case.



Let us consider an atom in the cavity, while participating in the a-c maser operation and being in the superposition

$$|\psi(t)\rangle = \ldots . |a\rangle\, e^{-iE_a t/\hbar} + \ldots . |c\rangle\, e^{-iE_c t/\hbar}$$

Its magnetic dipole moment coupling to the cavity mode, depends on the phase difference:

$$\langle M\rangle_t = \langle\psi(t)|M_{op}|\psi(t)\rangle = M_0 \cos(\underbrace{\omega_{ac}}_{(E_c-E_a)/\hbar} t + \delta),$$

δ standing for the relative phase of the coefficients represented by the dots. Now take an elastic collision of this atom with a non-masing d atom which is also present in large quantities. On the basis of the superposition principle, one may consider separately the collisions of the a and c parts with the d atom, which multiplies the first term by $S_{ad\to ad}$. By the same collision the c term is multiplied by $S_{cd\to cd}$. This induces an additional phase difference $\Delta\delta$ and an associated frequency shift $d\delta/dt$ proportional to the number of collisions with d atoms per second, i.e.

$$\sim n_d \arg\left[S_{ad\to ad}\, S^*_{cd\to cd}\right]$$

By a similar reasoning the collision of <u>two</u> masing atoms leads to a shift

$$\sim n_a \arg\left[S_{aa\to aa}\, S^*_{ca\to ca}\right] + n_c \arg\left[S_{ac\to ac}\, S^*_{cc\to cc}\right]$$

(Notes: 1) In deriving such frequency shifts from the quantum Boltzmann equation one gets slightly different expressions, but these formulas are sufficiently accurate in practice. 2) The second masing atom, although in a coherent superposition, is to be taken into account as an incoherent combination of a and c.)

Now, neglecting the hf interaction during the collision one can again apply

the superposition principle and consider for instance the aa state, prepared by the hyperfine coupling at larger distances, as a superposition of S = 0 and S = 1 states, each of the two behaving independently at short distances. One then finds the single-masing-atom contribution to be zero, while the two SS^* phases in the two-masing-atom shift are just equal and opposite, making the frequency shift $\sim n_a - n_c$. There is another frequency shift which is also proportional to $n_a - n_c$, namely the cavity pulling shift due to the detuning of the cavity. This makes it possible to tune the cavity such that these two frequency shifts cancel. That is, however, no longer true when the *effect of the hf interaction during collisions is taken into account*. Due to the fact that

$$[V^{hf},\ V^{exch}] \neq 0$$

a much more complicated calculation is required. We have done a rigorous calculation with the coupled-channels method and as an alternative a first-order treatment of the weak hyperfine interaction. The way the latter has to be carried out is a subtle question by itself, because the perturbation V^{hf} is of course not localized as a function of interatomic distance, so that the Born integral does not converge. It turns out, however, that one can adapt the Born treatment suitably. That this should be possible in principle is already clear intuitively from the fact that the above commutator does represent a localized perturbation. The result turns out to be that the above cancellation does not occur. This introduces an $n_a + n_c$ term. The single-masing-atom contributions no longer vanish, thus introducing n_d and n_b terms (also b atoms are produced within the cavity in H + H collisions). These additional hyperfine induced terms are not compensated by the cavity tuning procedure and are of significance in limiting the improvement in frequency stability attainable by cooling the H-maser (See contribution by Koelman et al.).

2. **Relaxation in degeneracy regime**

We now turn from the above phenomena associated with the coherence of hyperfine states to rate-like quantities. The relaxation of H↑↑ and D↑↑ in magnetic traps has been studied in previous papers[6-9] without taking degeneracy into account. Extending the calculations in this respect is of interest since a significant modification of the rates can be used as a signal of degeneracy. The present calculation proceeds along the lines of Kagan et al.[10] but is applied to two-body relaxation in an arbitrarily shaped potential well and extended to fermions.

We start with the number of transitions per second as given by Fermi's golden rule:

$$W = \frac{2\pi}{\hbar} \sum_{i,f} \rho_i \left| \langle f|V|i\rangle \right|^2 \delta(E_f - E_i)$$

$$= \int_{-\infty}^{\infty} dt \sum_{i,f} \rho_i \ \langle i|V(0)|f\rangle\langle f|V(t)|i\rangle$$

with $V(t) = \exp iH_0 t\ V \exp -iH_0 t$, V being the electronic magnetic dipolar interaction. In second quantization

$$V = \frac{1}{2} \iint d^3r_1 d^3r_2\ \psi^\dagger(r_1)\psi^\dagger(r_2)\ V^{dip}(r_1,r_2)\ \psi(r_1)\psi(r_2)$$

in which in self-evident notation

$$V^{dip} = c^{ee} \sum_{\mu} (-)^{\mu} (S_1 S_2)_{2,-\mu} \, Y_{2\mu}(\hat{r_1 - r_2}) / |r_1 - r_2|^3 .$$

Denoting the final two-atom states as $|p_1'\alpha, p_2'\beta\rangle$, α and β denoting spin states, we have

$$\sum_f \langle i|V(0)|f\rangle\langle f|V(t)|i\rangle = \tfrac{1}{4} L^{-3} (c^{ee})^2 \sum_{\substack{p_1'\alpha \\ p_2'\beta}} \iiiint d^3r_1 d^3r_2 d^3r_1' d^3r_2'$$

$$\langle i|\psi^\dagger(r_1,0)\psi^\dagger(r_2,0)\psi(r_2',t)\psi(r_1',t)|i\rangle \qquad (1)$$

$$(S_1S_2)^{\alpha\beta,dd*}_{2,-\mu} (S_1S_2)^{\alpha\beta,dd}_{2,-\mu} \frac{Y^*_{2\mu}(\hat{r_1 - r_2})}{|r_1 - r_2|^3} \frac{Y_{2\mu}(\hat{r_1' - r_2'})}{|r_1' - r_2'|^3}$$

$$\exp\left\{ip_1'(r_1 - r_1') + ip_2'(r_2 - r_2')\right\} \exp\left\{i(p_1'^2/2m + p_2'^2/2m - \Delta_{\alpha\beta})t\right\}$$

in which the spin matrix elements are to be calculated in the local B-field at r_1 and r_2, respectively, while Δ is the inelasticity associated with the process $dd \to \alpha\beta$ (for $D\uparrow\uparrow$: $\zeta\zeta \to \alpha\beta$). Anticipating the further approximations

$$\Delta_{\alpha\beta} \simeq \Delta_{\alpha\beta}(r_1) \simeq \Delta_{\alpha\beta}(r_1').$$

In the spirit of the WKB approximation for non-condensate atoms, the free atom states are locally eigenstates of H_0. To simplify (1) we introduce some appromations which are based on the slow variation of the correlator with the positions and the time. The slow variation of the correlator over the range of the dipole interaction leads us to take for bosons

$$r_1 \simeq r_2, \; r_1' \simeq r_2'$$

in the correlator. For fermions, according to the Pauli principle

$$\psi(r',t)\psi(r',t) = 0,$$

so that we switch to the first-order Taylor term, giving rise to

$$(\psi^\dagger \nabla\psi^\dagger)_{r,0} \, (\psi\nabla\psi)_{r',t}$$

Also for bosons one can add such higher-order terms if desirable.
The remaining variations of the correlator with position (over $\hbar/\sqrt{2m\Delta}$) and time (over $\hbar/\Delta$) being also slow, we take in addition $r \simeq r'$ and $t \simeq 0$. For bosons we split $\psi(r)$ in a condensate wave function $\psi_0(r)$ and a non-condensate part $\psi'(r)$ and calculate the correlator for the ideal gas:

$$\langle\psi^\dagger\psi^\dagger\psi\psi\rangle = \left\{n_0^2 + 4\, n_0 n' + 2\, n'^2\right\}_{r,T}$$

making use of

$$\langle\psi'^\dagger\psi'^\dagger\psi'\psi'\rangle = 2\,\langle\psi'^\dagger\psi'\rangle\,\langle\psi'^\dagger\psi'\rangle \equiv 2\, n'^2$$

In the case of fermions we find

$$\langle\psi^\dagger\nabla\psi^\dagger\;\psi\nabla\psi\rangle = -\,\langle\psi^\dagger\psi\rangle\,\langle\nabla\psi^\dagger\nabla\psi\rangle + \tfrac{1}{4}\,\langle\nabla(\psi^\dagger\psi)\rangle^2$$

$$= \left\{ -\frac{2m}{\hbar^2} n.t + \frac{1}{4} (\nabla n)^2 \right\}_{r,T} \simeq -\frac{2m}{\hbar^2} n^2 \langle E_{kin} \rangle_{r,T}$$

in which t is the thermally averaged kinetic energy density.
In total we find

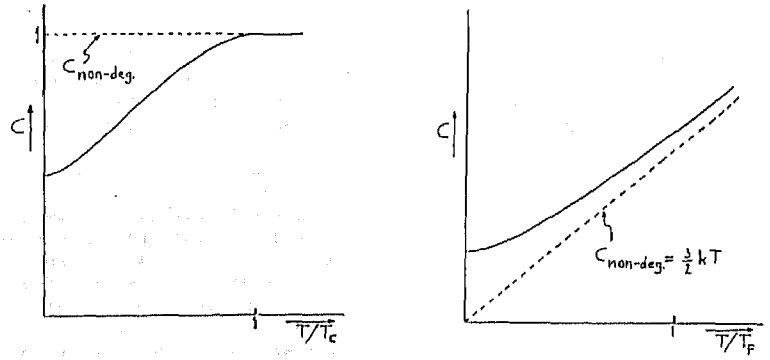
$$W = \int d^3r \; W_0(r) \; C(r,T)$$

where $W_0 \sim n^2$ is different for bosons and fermions, but common for the degenerate and non-degenerate regimes, while

$$C(r,T) = \frac{1}{2n^2} \left\{ n_0^2 + 4 n_0 n' + 2 n'^2 \right\}_{r,T} \qquad \text{(Bose)}$$

$$C(r,T) = \langle E_{kin} \rangle_{r,T} \qquad \text{(Fermi)}$$

As an illustration we give the temperature dependence of C for the special case of a uniform system of H↑‡ atoms as a function of T/T_c:



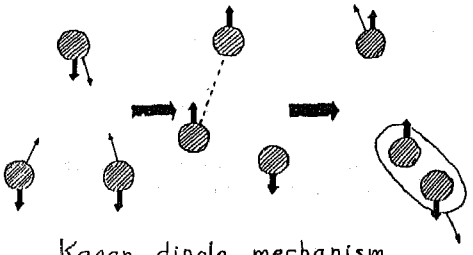
Note that the discontinuity at $T = T_c$ is in the second derivative. The result is a decrease of the relaxation rate upon the appearance of a Bose-condensate. Like the three-body recombination in H↓‡[10], this can be used as a signal of the phase transition.
Likewise, we give C for the case of D↑‡ as a function of T/T_F. Now, an increase with respect to the non-degenerate situation is predicted, due to the fact that the Pauli principle forbids the occupation of single-particle states with low kinetic energy by more than one atom.
So far for the calculations of dipolar relaxation of bosonic and fermionic low-field seekers. Both cases of bosons and fermions are dealt with more extensively in two contributed papers.

3. H↓‡ in strong magnetic fields

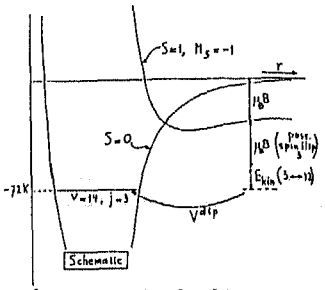
For physical interest as well as to explore the possibilities of producing the Bose-Einstein consensed state in H↓↓ at high densities it is of interest to clarify the mechanism of the thresholdless recombination process $H + H + H \rightarrow H_2 + H$. The aim is to resolve the existing discrepancies with experiment in the bulk and adsorbed gases at lower fields and subsequently to explore the possible existence of a B-field "window" with a

low enough rate at stronger fields. The nature of the discrepancy with experiment being similar with respect to B-dependence in the 2D and 3D gases (calculated values strongly increasing instead of slowly decreasing), our strategy is to assume a common explanation and to investigate primarily the volume recombination which is amenable to more rigorous calculation. From the point of view of trying to find a B-field "window" it is of course of interest to dispose of a reliable theory in B-field regions where the rates are small.

In 1981 Kagan et al.[11] described a dipolar mechanism for three-body recombination: two H-atoms with initial electron spins parallel collide with a third atom, which combines the roles of

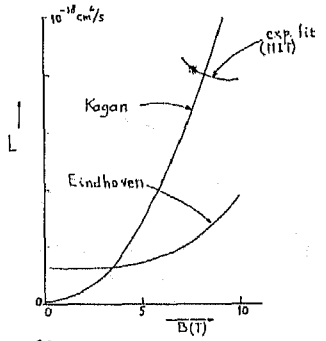


Kagan dipole mechanism



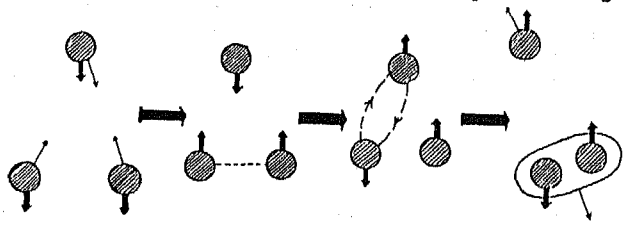
introducing an antiparallel spin component by the magnetic field difference at the place of the first two atoms and carrying off energy and momentum. The molecular binding energy ΔE_{vJ} released is spent to the single or double spin flip (1sf or 2sf) as well as to the final atom-molecule kinetic energy:

$$\Delta E_{vJ} = (2)\mu_B B + p_3^2/\frac{4}{3}m_H$$



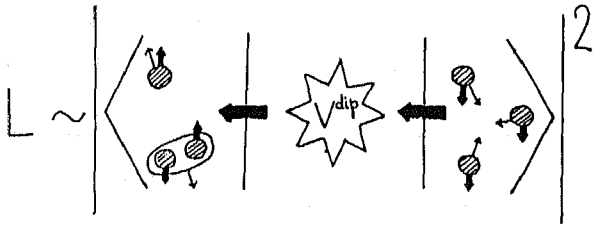
For each vJ level this gives rise to a cut-off field $B_{max}(vJ,isf)$. With some approximations, among which a free atom motion in the final state, Kagan et al.[11] predicted a recombination rate according to the figure: a strongly rising field-dependence. As pointed out in the foregoing this prediction turned out to differ markedly from the slowly decreasing experimental behavior obtained by the MIT, Amsterdam/Harvard and Turku groups (See e.g. the MIT experimental fit)

A possible additional contribution is the dipole-exchange mechanism.



Dipole - exchange mechanism

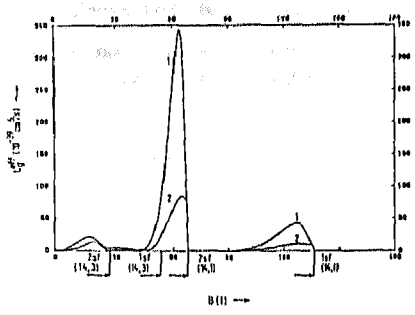
Now the two recombining atoms undergo a mutual dipole interaction, their spins remaining parallel, followed by an exchange interaction with the third atom which again introduces an antiparallel spin component. Whereas Kagan et al.[11] neglected this contribution, we found it on the basis of a model calculation to be very hopeful both with respect to B-dependence and magnitude. The uncertainties inherent in the model led us to carry out a more rigorous calculation, starting from the expression



with the central interactions to be included in principle exactly in the initial and final states. In view of the intricacies of such a calculation we decided to follow a stepwise approach, introducing the complications one by one. By a Faddeev type calculation we calculated the initial state rigorously. The above figure shows our result with all (in)elastic atom-molecule interactions coupling rotational/vibrational states taken into account in the final state. This calculation which is rigorous except for rearrangement (the exchange of the single atom and one of the bound atoms) is the present state of the art. It is already possible to conclude in this stage that the original agreement in magnitude of the rate between the Kagan result and experiment was accidental: At B =7.6T, for instance, our calculated value is roughly a factor of 4 lower than experiment. That makes it probable that rearrangement, including the dipole-exchange process, is responsible for the major part of the experimental rate at these fields. Such a calculation is in progress. It is based on the variational principle

$$\langle \delta\psi_S | H - E | \psi_S \rangle = 0$$

in which ψ_S is a (symmetric) sum of three terms each of which has an atom pair in an arbitrary bound state (Variation after symmetrization; the previous method can be characterized as symmetrization after variation). This resonating group method therefore includes rearrangement and all (in)elastic couplings, but excludes the possibility of a three-body continuum part.



In view of recent measurements at very strong magnetic fields by the Harvard group and because it illustrates some of the underlying physics, we extended the above calculations without rearrangement to higher B. Curve 1 is the result according to Kagan's approximations. Curve 2 is our most rigorous calculation which still neglects rearrangement. Also indicated are the field ranges for single and double spin flip. The repetitive structure as a function of B (double spin flip followed by single spin flip) is clearly

seen. The most conspicuous features of this figure are: 1) The $(B_{max} - B)^{j+1/2}$ downward right-hand slope of each of the maxima due to the centrigugal barrier felt by the final atom. 2) The left-hand slope due to the decreasing overlap of the growing final momentum p_3 with the small momenta in the initial state. 3) The decrease of our values compared to Kagan's by a factor of about 4 outside the narrow field regions close to the cut-off fields, mainly due to the inclusion of correlations in the final state, which keeps the third atom out of close distance from atoms 1,2. Clearly, this reduction is expected to be less close to the cut-off fields: The centrifugal barrier in the final state then by itself already keeps the final atom out of the forbidden part of space, which makes the Kagan model an excellent approximation. In the same narrow regions just left of the cut-off fields we expect the dipole-exchange process too to be negligible due to the same centrifugal effect. We repeat that from the point of view of Bose-condensation one is primarily interested in field strengths where the rate is small. In this respect it is very interesting to see what a calculation including rearrangement will predict.

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