V. ATOMIC RESONANCE AND SCATTERING[*]

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RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH

Our group is concerned with atomic structure and with interactions between atoms.

Experiments are in progress to determine the magnetic moments of the proton and deuteron to high precision, and to obtain absolute values of nuclear shielding constants in some simple molecules. The experiments are carried out with a hydrogen maser and an associated magnet of unusually high homogeneity and stability.

Our investigations of atomic interactions are centered on a new technique that has recently been developed within our group — differential spin-exchange scattering. During last fall, we obtained the first data of high quality; the initial results indicate clearly that spin-exchange scattering will indeed be a fruitful technique.

Recently, we initiated investigation of a new superconducting device that may permit detection of individual hydrogen atoms, and possibly infrared photons. If successful, the detector will have important applications in scattering, and possibly in other areas of physics.

D. Kleppner

A. DIFFERENTIAL SPIN-EXCHANGE SCATTERING

The object of these studies is to investigate spin-dependent interactions in atomic systems. The technique involves scattering in a crossed atomic beam scattering apparatus in which the incident and scattered beam are spin-analyzed. Since the apparatus was described in Quarterly Progress Report No. 84 (page 27), the present report will be concerned chiefly with interpretation of the experiments.

Consider a collision between two single electron atoms such as two alkalis. (Because of the low collision energy (<1 eV), we assume that only the valence electron plays a dynamical role.) The atoms are influenced by the long-range spin-independent Van der Waals attraction, and by an exchange force that is attractive in the singlet state and repulsive in the triplet state. When the exchange potential is small relative to the excitation energy of the atoms it has the same magnitude (but opposite sign) and the potential may be written

$$V_{1,3} = -\frac{C}{r^6} \mp \frac{\delta V(r)}{2}.$$

Each spin component of the incident wave propagates with its individual scattering amplitude, and interference between the singlet and triplet amplitude can result in spin exchange. Our experiments give a direct measure of this interference, and thus yield detailed information on the singlet and triplet potentials, V_1 and V_3.

For concreteness, we shall consider the collision of two atoms with spins opposed. The initial spin state is

$$\chi_i = |\uparrow\downarrow\rangle = \frac{1}{\sqrt{2}}|1,0\rangle + \frac{1}{\sqrt{2}}|0,0\rangle, \tag{1}$$

where $|s, m_s\rangle$ is a state function of the coupled system. The scattered wave is

$$\chi_s = \frac{1}{\sqrt{2}} f_3(\theta)|1,0\rangle + \frac{1}{\sqrt{2}} f_1(\theta)|0,0\rangle \tag{2}$$

which, in the independent spin representation, is

$$\chi_s = \frac{1}{2}(f_3+f_1)|\uparrow\downarrow\rangle + \frac{1}{2}(f_3-f_1)|\downarrow\uparrow\rangle. \tag{3}$$

The first term corresponds to an atom scattered in the original spin state, and the second describes a state that has undergone spin exchange.

If the atoms approach with parallel spins, then the scattering is described completely by f_3, and no exchange can occur. Since our targets are unpolarized, an average must be done over all orientations, with the result

$$\left(\frac{d\sigma}{d\Omega}\right)_{||} = \frac{1}{8}|f_3+f_1|^2 + \frac{1}{2}|f_3|^2$$
$$\left(\frac{d\sigma}{d\Omega}\right)_x = \frac{1}{8}|f_3-f_1|^2. \tag{4}$$

The subscripts x and ||, respectively, indicate no spin-exchange and spin exchange.

The differential cross section summed over spin states is

$$\frac{d\sigma}{d\Omega} = \left(\frac{d\sigma}{d\Omega}\right)_{||} + \left(\frac{d\sigma}{d\Omega}\right)_x = \frac{3}{4}|f_3|^2 + \frac{1}{4}|f_1|^2. \tag{5}$$

The probability of exchange, P_{ex}, a useful experimental quantity, is

$$P_{ex} = \left(\frac{d\sigma}{d\Omega}\right)_x \Big/ \left(\frac{d\sigma}{d\Omega}\right) = \frac{|f_3-f_1|^2}{6|f_3|^2 + 2|f_1|^2}. \tag{6}$$

The expression (6) must be modified to account for nuclear spin effects. It has been shown[1] that the result of this is to multiply P_{ex} by a factor $m(I) = 1 - 2I/(2I+1)^2$, where I is the nuclear spin of the observed atom, with $m(I)$ ranging between 0.75 and 1 as I varies between $1/2$ and ∞. If we let $x = |f_1| / |f_3|$, and represent the phase angle between f_3 and f_1 by δ, then the probability of exchange measured by the apparatus is

$$P_{mx} = m(I)\, P_{ex} = m(I)\, \frac{1 + x^2 - 2x \cos \delta}{6 + 2x^2}. \tag{7}$$

Figure V-1 is a plot of allowed values of P_{ex} for all possible values of x and δ. As we shall show, this plot can give immediate insight into spin-exchange measurements.
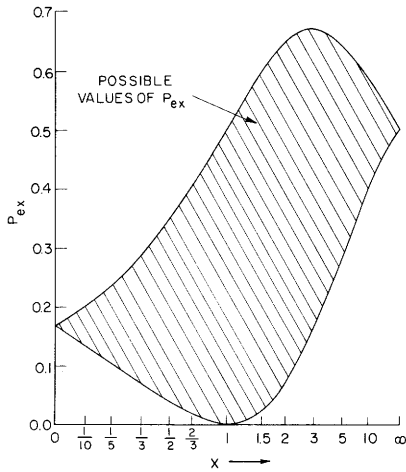

Fig. V-1. Probability of spin exchange vs x, where $x = |f_1| / |f_3|$.

If the singlet and triplet potentials, V_1 and V_3, respectively, are known, their respective phase shifts can be calculated from the JWKB approximation.[2]

$$\eta_\ell^{1,3} = \frac{\pi}{2}\left(\ell + \frac{1}{2}\right) - kr_0 + k \int_{r_0}^{\infty} \left\{ \left(1 - \frac{V}{E} - \frac{\left(\ell + \frac{1}{2}\right)^2}{k^2 r^2} \right)^{1/2} - 1 \right\} dr, \tag{8}$$

where r_0 gives the zero of the integrand, k is the momentum, and E is the kinetic energy. (Atomic units are used throughout.) The scattering amplitudes are given by the well-known formula

$$f_{1,3}(\theta) = \frac{-i}{2k} \sum_{\ell=0}^{\infty} (2\ell+1) \left\{ e^{2i\eta_\ell} - 1 \right\} P_\ell(\cos \theta). \tag{9}$$

In order to obtain accurate results, the expressions (8) and (9) must be evaluated

numerically. Work on this is in progress. For certain potentials many features of the scattering can be found analytically, however. In particular, if the potential is of the 6-s form, $V = -C/r^6 + D/r^s$, then the phase can be found analytically,[3] and by converting Eq. 9 into an integral and evaluating by the method of stationary phase,[2] an approximate value of $f(\theta)$ is obtained.

Of particular importance is the rainbow angle, θ_r, which corresponds classically to a singularity in f, because of a stationary point in the deflection function vs impact parameter. This angle is related to the well depth, V_{min}, by

$$\frac{V_{min}}{E} = G(s)\, \theta_r, \tag{10}$$

where $G(s)$ is a slowly varying function of s. $G(s)$ is given by

$$G(s) = \left(\frac{16}{15\pi}\right)^{s/(s-6)} \left(\frac{\pi s}{2^{s+1}} \frac{s!}{\left(\frac{s}{2}!\right)^2}\right)^{6/(s-6)}. \tag{11}$$

A few values of $G(s)$ are

s	8	10	12
G(s)	0.5392	0.5105	0.4902

This result is substantially modified by the semiclassical treatment of (9), although it is still useful in an initial attempt to find V_{min}. From the results of Ford and Wheeler,[2] it can be shown that the maximum in f no longer occurs at θ_r, but at

$$\theta_{max} = \theta_r - \left[\left(60\left(\frac{2}{5}\right)\right)^{4/3}\right]^{1/3} \left(\frac{3\pi}{16}\mu k^4 C\right)^{-1/9} \times 1.018\, \theta_r^{4/9}. \tag{12}$$

Here μ is the reduced mass in atomic units.

This implicit expression for θ_r can yield a reliable result for the well depth. Numerical evaluation is required, however, to fit subsidiary maxima of $d\sigma/d\Omega$.

As an example of how these ideas can be applied, we present in Fig. V-2 results for spin-exchange scattering of Na from Cs. The relative velocity was 1.30×10^5 cm/sec.

The pronounced dip in P_{ex} at 180 mrad apparently is attributable to rainbow scattering in the triplet state. Evidence for this is that the minimum in P_{mx} is 0.14, which is in good agreement with the value $0.813 \times (1/6) = 0.137$ predicted from (7) or Fig. V-1. Beyond θ_r, f_3 rapidly decreases, and P_{ex} increases. The maximum observed value,
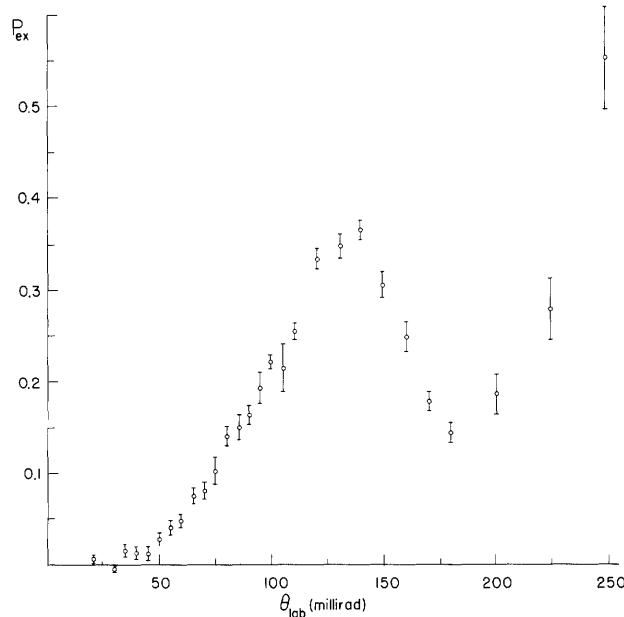
Fig. V-2. Probability of exchange vs scattering angle for
Na-Cs collisions. The relative velocity was
1.30×10^5 cm/sec.

0.55 at 250 mrad is close to the maximum possible value, 0.54, which occurs when $f_1 = -3f_3$.

By tentatively identifying 180 mrad (or 205 mrad in the center-of-mass system) as θ_{max} for the triplet rainbow, we find from (12) that θ_r = 259 mrad. (We have used the result of Buck and Pauly[4] for the Van der Waals constant.)

Near the triplet rainbow, $d\sigma/d\Omega$ is dominated by triplet scattering and the subsidiary maxima can be used to find the potential parameter, s. By using the numerical results of Hundhausen and Pauly,[5] we obtain the following results for the region of the potential minimum:

$$V_{min} = 9.8 \pm 1.0 \times 10^{-4} \text{ a. u.}$$

$$r_{min} = 8.55 \text{ a}_o.$$

Furthermore, by fitting the data to numerically calculated positions of the subsidiary maxima and minima,[5] we are able to infer that a 6-8 potential gives a better fit than a 6-12 potential. This tells us that the term D/r^8 is a good fit to $\delta V(r)/2$ in the region close to 8.55 a_o, when D is adjusted to give the proper depth of the potential well.

Let us now turn our attention to the information that is contained in the behavior of $P_{ex}(\theta)$ at angles less than 130 mrad. We can infer the quantity $\delta(\theta)$ from P_{ex} by using Eq. 7 and assuming that x is close to 1, an assumption that is justified by the fact that

$P_{ex} \approx 0.45$ at 130 mrad, which requires $x \geqslant 0.80$. Scattering at these small angles corresponds to impact parameters significantly greater than $r_{min} = 8.55 \, a_o$, a region in which $C/r^6 \gg \delta V(r)$. We are therefore justified in assuming that all of the deflection arises from the Van der Waals term in the potential, and this permits us to determine the impact parameter uniquely from the scattering angle. The result is that we can find the function $\delta(b)$ experimentally.

Theoretically, $\delta(b)$ is given by

$$\delta(b) = \frac{1}{\hbar v} \int_{collision} \delta V(r) \, d\ell. \tag{13}$$

If we assume that $\delta V(r) \sim A/r^n$, then

$$\delta(b) = \frac{\sqrt{\pi}}{2} \frac{\Gamma\left(\frac{n-1}{2}\right)}{\Gamma\left(\frac{s}{2}\right)} \frac{A}{\hbar v b^{n-1}} = \frac{\sqrt{\pi}}{2} \frac{\Gamma\left(\frac{n-1}{2}\right)}{\Gamma\left(\frac{s}{2}\right)} \frac{b}{\hbar v} \delta V(b).$$

Comparison of this expression with the data shows that $n \approx 12$, which permits us to obtain $\delta V(b)$ directly from $\delta(b)$. Experimental points for $\delta V(b)$ obtained in this manner are plotted in Fig. V-3.
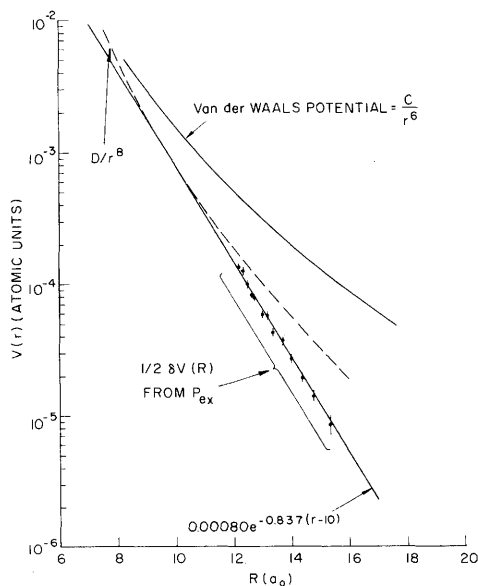

Fig. V-3.

Na-Cs triplet potential vs internuclear separation. The curve for D/r^8 is calculated from rainbow data. The data points shown for large internuclear separations are derived from the observed exchange probability. For comparison, the Van der Waals potential is also shown.

While neither the D/r^8 term, which fits $\delta V/2$ in the region near the rainbow, nor the $A/2r^{12}$ term, which fits $\delta V/2$ at larger impact parameters, will give a good fit to $\delta V/2$ when extrapolated to the other region, it is clear from Fig. V-3 that the expression

$$\frac{\delta V(r)}{2} = 0.00080 \, e^{0.87(10-r)} \qquad \text{atomic units}$$

fits it well in both regions. This is a reasonable result, for we expect δV to have an exponential dependence on r, since it results from an overlap of the two electron clouds, both of which have exponential dependence on r. This fine agreement between the two methods of determining $\delta V/2$ is a strong indication that the techniques described in this report, while only approximate, are reasonably accurate. Work involving an exact computer calculation of $\left(\dfrac{d\sigma}{d\Omega}\right)$ and $P_{ex}(\theta)$ from various potentials will be a final check on the results.

<div align="right">D. E. Pritchard, G. N. Carter, F. Y. Chu, D. Kleppner</div>

References

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