PHYSICAL REVIEW LETTERS

VOLUME 18 — 6 MARCH 1967 — NUMBER 10

STIMULATED EMISSION AND Rb SPIN-EXCHANGE CROSS SECTION

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(Received 5 January 1966)

During the last few years several measurements of spin-exchange cross sections for alkali-alkali collisions have been reported.[1-8] Special emphasis has been placed on Rb-Rb collisions, and in general, the results are scattered between values of 1 and 8×10^{-14} cm^2; the most reliable measurements lie in the lower values.[3,4] In most of the measurements made, a relaxation rate for the populations of specified levels is measured and this is compared with $n\bar{v}_r\sigma$, where σ is the relaxation cross section caused by spin exchange and $\bar{v}_r$ is the atoms' average relative velocity. The density n is the most difficult parameter to obtain for a specific experimental situation. In general it is found that the vapor pressure inside a given system, which is normally made of glass, depends very strongly on the history and geometrical shape of the container and rubidium reservoir. Consequently, errors even of an order of magnitude are possible in the determination of the density from vapor pressure tables which themselves are not too reliable. In the most accurate measurements, the density is determined through an interferometric method in which one measures the amount of resonance light absorbed by the sample under study.[3,8] That method depends on a knowledge of the value of the absorption coefficient for resonance radiation.

In this article an experiment is described in which the rubidium cross section is obtained without a knowledge of the rubidium density. The technique uses the phenomenon of stimulated emission; in practice, the emission is obtained after a microwave resonant pulse, whose length is such that the driving phase angle is less than $\frac{1}{2}\pi$. The behavior of atomic systems in that case has been described in a semiclassical manner by Bloom.[9] In his calculation it is assumed implicitly that the relaxation rates $\gamma_1(=1/T_1)$ and $\gamma_2(=1/T_2)$, which characterize, respectively, the decay of the populations and the decay of the oscillating moment, are equal. A density matrix solution of the same problem gave the result obtained by Bloom only when γ_1 was made equal to γ_2.

The experimental arrangement used is essentially a rubidium maser.[10] It is shown in Fig. 1 with its associated electronics. In this arrangement the dc magnetic field is parallel to the z components of the rf field in the cavity and the transition involving levels $F=2$, $M_F=0$ and $F=1$, $M_F=0$ is studied. The atomic system is prepared by a strong pulse of light which produces an inversion of the population. The applied pulse of light is long enough to produce equilibrium and we define $\Delta_0=\rho_{33}-\rho_{77}$, where ρ_{33} and ρ_{77} are diagonal elements of the density matrix representing the rubidium atoms. The levels are numbered from high to low energy; the value of Δ_0 is positive and is given by

$$\Delta_0=\Gamma'(5\Gamma'+8), \tag{1}$$

where Γ' is the ratio of the pumping rate to

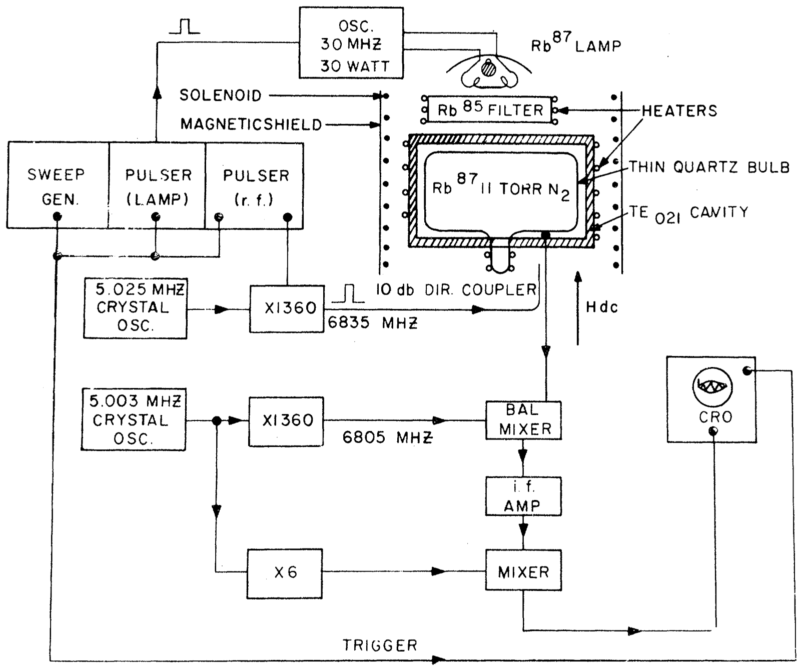

FIG 1. Experimental arrangement.

the relaxation rate γ_1. Γ is expressed in photons per atom per second[11] and following Balling, Hanson, and Pipkin[2] we average its value over the volume occupied by the atoms. A microwave resonant pulse is applied immediately after the light pulse and induces the atoms into a radiant state. In the following lines we reproduce the results obtained by Bloom, with a few changes in notation in order to make the equations easier to compare with our experiments. The form of the ringing that follows the microwave pulse is given by

$$P=P_m e^{-2\gamma t}\operatorname{sech}\left[-\frac{\Delta_0 k}{\gamma}\left(1-e^{-\gamma t}\right)-\ln\tan\frac{\theta_p}{2}\right], \quad (2)$$

where P is the power output, γ the relaxation rate, and θ_p the driving phase angle of the microwave pulse. For a short rf pulse, the angle θ_p determines the population difference through $\Delta=\Delta_0\cos\theta_p$. The parameter k is given by $k=(4\pi Q_l\eta\mu_0^2/V_c\hbar)nV_b$, where V_c is the volume of the cavity, V_b is the volume of the storage cell, η is the filling factor, μ_0 is the Bohr magneton, and Q_l is the loaded quality factor of the cavity. P_m is given by $P_m=\frac{1}{2}h\nu Nk\Delta_0^2$, where ν is the resonant frequency and N is the total number of atoms in the bulb.

Equation (2) predicts a delayed surge of power with a maximum at a time t_M given by the following equation:

$$1=\exp(-\gamma t_M)\tanh\left\{-\frac{\Delta_0 k}{\gamma}[1-\exp(\gamma t_M)]-\ln\tan\frac{\theta_p}{2}\right\}. \quad (3)$$

We have observed the delayed peak with the arrangement of Fig. 1. A typical result is shown in Fig. 2. The experimental data agreed well with the predictions of Eq. (5) if precautions were taken in evaluating θ_p. In the experiment it was found more accurate to assess the value of θ_p from the amplitude of the signal immediately after the microwave pulse rather than from the pulse length which was the variable parameter. This was primarily due to the dependence of the pulse amplitude on the

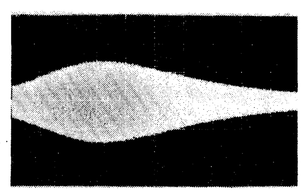

FIG. 2. Typical stimulated emission signal after a microwave resonant pulse whose driving phase angle is less than $\frac{1}{2}\pi$.

pulse length in the case of short pulses. Just after the microwave pulse the amplitude of the induced signal is proportional to $\sin\theta_p$; consequently a knowledge of the amplitude for $\theta_p = 90°$ permits the evaluation of θ_p for other amplitudes.

In order to determine the spin-exchange cross section, we have used the above phenomenon in the following way: From Eq. (3), t_M is a function of θ_p and tends to 0 for a given pulse length such that

$$(\cos\theta_p)_{t_M=0}{}^{-1} = \frac{\Delta_0 k}{\gamma}. \tag{4}$$

In practice, the value of θ_p for $t_M = 0$ can be obtained from a plot of t_M vs θ_p. From the value of the parameter k defined earlier, one can write

$$\frac{\Delta_0 k}{\gamma} = \frac{4\pi\mu_0{}^2}{\hbar}\frac{Q_l \eta n V_b}{V_c}\frac{\Delta_0}{\gamma}. \tag{5}$$

It was found experimentally that the relaxation rate γ_1 was mostly due to spin-exchange interactions, with a few percent due to collisions between rubidium and nitrogen, which is used as a buffer gas. It was found also that as the rubidium pressure is raised, γ_2—initially larger than γ_1 because of large contribution from the buffer gas—increases at a lower rate than γ_1. This is because, in spin-exchange processes, T_2 is longer than T_1.[12] At a temperature of about 70°C both relaxation rates become equal. Along with this reasoning, one can write $1/T_1 \approx n\bar{v}_r \sigma = \gamma$ which, when replaced in Eq. (4), gives the value of σ as

$$\sigma = \frac{4\pi\mu_0{}^2}{\hbar\bar{v}_r} \times \frac{\Delta_0 Q_l \eta V_b}{V_c}(\cos\theta_p)_{t_M=0}. \tag{6}$$

All the parameters above are either obtained experimentally or are fundamental constants. The value of Γ which enters in the calculation of Δ_0 was determined by measuring the characteristic time taken to obtain equilibrium of the energy level populations at the beginning of the light pulse. The relaxation rate γ_1 was obtained from the stimulated emission signal after the microwave pulse in a method similar to the one used by Arditi and Carver.[13] The signal amplitude immediately after the microwave pulse plotted against the time elapsed between the end of the light pulse and the rf pulse gave a direct measure of the rate γ_1.

In these experiments, γ_2 was also measured from the decay of the stimulated emission. This was done in an experimental situation where the rf field produced by the atoms in the cavity had little influence on the decay itself. In that case the rf field in the cavity decays at the rate γ_2. Typical pulse lengths were 30 msec for the light and 50 μsec for the microwaves (90°).

The value of σ was determined for temperatures near 70°C, where $T_1 \approx T_2$, and was found equal to

$$\sigma = (2.02 \pm 0.20) \times 10^{-14}\ \text{cm}^2.$$

The possible error given above is the rms deviation of the various measurements made and represents primarily the scatter in the determination of $(\cos\theta_p)_{t_M=0}$. This value of σ is in rather good agreement with the value determined by Gibbs.[3] The absolute accuracy of the measurement reported here is, of course, a function of the absolute accuracy to which the various parameters of Eq. (6) can be determined. The filling factor η is probably the most uncertain parameter; in the present case the shape of the storage bulb used did not permit its exact calculation. The value of η was estimated from its maximum value for a storage cell completely filling the cavity and the ratio of the actual storage-cell volume to the total volume of the cavity. It is estimated, however, that in the present experiments, η was known to about 10% certainty. In the determination of Δ_0, it is observed that greater accuracy is obtained when Γ is large. In fact, due to the form of expression (1) and the size of Γ in these experiments, an error of 25% in Γ would be reflected by an error of only 7% in the value of Δ_0. The use of a bright lamp characterized by a large Γ thus increases the accuracy to which Δ_0 is known and also diminishes the effect of nonuniform illumination that could exist at the densities at which the measurements were made. Because of these effects the absolute accuracy on σ is expected to be somewhat less than the accuracy that was quoted above, which reflects the reproducibility of the measurements. Experiments with a vacuum-tight cavity illuminated from both ends by bright lamps should increase the accuracy of σ because η and Δ_0 would be better known.

The above method of measurements can be extended to several other atomic systems. An interesting case would be the determination of the hydrogen spin-exchange cross section in hydrogen-hydrogen collisions. This could be done in the hydrogen maser.[14] In that case, since the population inversion is obtained through magnetic selection, the atomic beam would be pulsed. This experiment would be of basic importance because a large amount of theoretical work has been done on spin-exchange interactions between hydrogen atoms and because little experimental data are available.

The author has benefited from several discussions with Dr. R. F. Lacey and Professor R. Novick. The work described above has been funded by the U. S. Electronics Command, Fort Monmouth, New Jersey.

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STRONG-COUPLING SUPERCONDUCTIVITY IN GALLIUM

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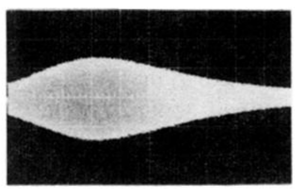
(Received 12 January 1967; revised manuscript received 10 February 1967)

Ordinary gallium has a superconducting energy gap $\Delta_0 = 0.16$ mV and is a weak-coupling superconductor ($2\Delta/k_BT_c = 3.5$). We have prepared Ga films which exhibit three distinct energy gaps: $\Delta_1 = 1.03$ mV, $\Delta_2 = 1.38$ mV, and $\Delta_3 = 1.53$ mV. The corresponding values of $2\Delta/k_BT_c$ are substantially larger than the BCS value[1] 3.5, indicating strong-coupling superconductivity. We believe that the three energy gaps are those of three high-T_c modifications of Ga present in the films. The high Δ's can be accounted for by an increase in N, the density of electronic states at the Fermi surface, above that of ordinary gallium, which has the lowest value of N of all superconductive elements.[2]

The Ga films were evaporated from an alumina-coated tungsten filament at the rate of 70 Å/sec onto a microscope slide at room temperature. An oxygen pressure of 10^{-4} mm Hg was maintained during the evaporation. The oxygen provides nucleation centers for the film to grow in the form of extremely small particles.[3] The small particle size is favorable for the formation of high-T_c phases of gallium.[4] The T_c of the films did not change significantly upon storing at room temperature for several days. Emission spectroscopy of the films showed a metallic impurity content of less than 0.1%.

The energy gaps were determined by means of tunneling measurements using Al-Al_xO_y-Ga, Sn-Sn_xO_y-Ga, and Ga-Ga_xO_y-Pb junctions. The junctions were made of crossed strips of the two metals; the width of the Ga and Al strips was 1 mm, while that of the Pb or Sn strips was 0.5 mm. The Ga-Pb junctions were made by first evaporating a 500-Å-thick Ga film, letting it oxidize in air for 2 h, and then evaporating a 1500-Å-thick Pb film on top of the Ga. If the Ga film was evaporated on top of the Pb, the resulting junctions always had elec-



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