TABLE I. Correlation between the ultraviolet cutoffs (column 3) and double reflection intensities (column 5) in diamond. Weight of specimen (column 2), and type (column 4) are also given.

Specimen	Weight in mg	Cutoff A	Type	Double reflections
*R*25	2.9	2240	II	strong
*R*27	4.3	2930	I	weak
*R*28	3.5	2930	I	weak
*R*36	3.1	2250	II	strong
*R*40	5.0	2950	I	weak
*R*47	2.6	2250	II	strong
*R*51	4.1	2960	I	weak
*R*62	3.3	2990	I	weak
*R*69	4.4	2240	II	strong

{1,3,3} equivalent spots appearing with a periodicity of 60°. The camera was allowed to oscillate through 120° about the axis of rotation, so that two equivalent sets of spots appeared on the film. Intensities of these spots were found to be strongly correlated to the type of diamond as classified according to the ultraviolet absorption edges.

Many Type II diamonds showed some absorption at wavelengths above 2250 A, while some Type I specimens transmitted considerably below 3000 A. A full correlation between the double-reflection intensities of these intermediate cases and their ultraviolet absorption spectra necessitates quantitative intensity measurements; a modified camera will have to be constructed for this purpose. Therefore, the results of only a few typical Type I and Type II specimens are given now. Ultraviolet cutoffs for these specimens are given in Table I, and the corresponding double reflections are shown in Fig. 1. Two microphotometer traces, one for a Type I specimen (*R*27) and one for a Type II specimen (*R*47) are shown in Fig. 2. The difference is striking; while for Type I diamonds the double-reflection spots are almost invisible, they appear very strongly in the Type II photographs.

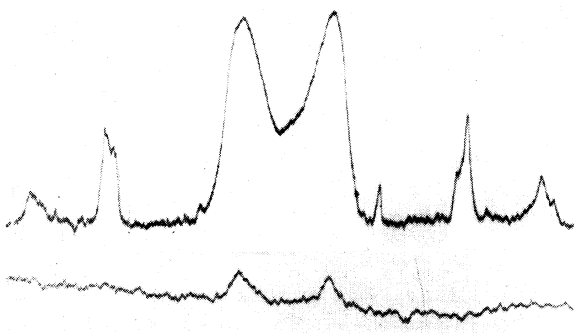

FIG. 2. Microphotometer traces for a Type I (*R*27) diamond—lower trace, and for a Type II specimen (*R*47)—upper trace.

In addition, the spots in Type II specimens do not appear exactly on the (2,2,2) line, as those of Type I specimens do, and some of them even show duplication.

It should be stressed that intensity ratios of the (2,2,2) reflections relative to the normal x-ray reflections remained constant for the specimens examined, within a factor of 1.5, which is a small variation compared to the variations in the intensities of the double-reflection spots.

In conclusion, the high intensities of the double reflections in Type II diamonds seem to imply that the perfect crystal blocks in these diamonds are bigger than those in Type I diamonds, whereas extension of the spots outside the (2,2,2) line might indicate that the mosaic blocks are badly aligned in Type II specimens.

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Slow Spin Relaxation of Optically Polarized Sodium Atoms*

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In order to obtain as narrow as possible paramagnetic resonance signals, it is of importance to investigate the conditions under which long relaxation times can be realized. In the present experiment on sodium atoms diffusing in argon gas, relaxation due to sodium-sodium collisions was minimized by employing very low sodium partial vapor pressures (about 10^{-7} mm Hg). While at lower pressures the argon is serving its function well to slow down relaxation by inhibiting wall diffusion, at about 10 cm Hg relaxation due to sodium-argon collisions becomes the decisive factor. Nevertheless it was possible to realize a relaxation time of 0.21 sec for a 1-liter spherical bulb filled with 3 cm argon. About 0.02 sec was found for a 0.1-liter, 40 cm argon sample. In carrying out the experiments, optical pumping by circularly polarized resonance radiation was used to create an orientation of the sodium atoms which then was monitored by measuring the transmission of the pumping radiation through the sample. By suddenly reversing a small axial magnetic field, the polarization of the atoms could be made to reverse too. From the decay rates of this inverted polarization under the combined effects of relaxation and continuing optical pumping, the experimental relaxation times were deduced. The strong signals obtained are indicative of available signal to noise ratios in future radio-frequency resonance reorientation experiments using the transmission monitoring technique. A theoretical analysis of the optical pumping process, including the dynamic aspects and allowing for collisions with argon that the sodium atoms undergo while in the excited state, was carried out and used to describe the experimental data.

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RECENT paramagnetic resonance *absorption* experiments[1,2] on free atoms diffusing back and forth in an inert buffer gas have created some interest in the spin relaxation processes in these systems. Various mechanisms have been discussed by Wittke and Dicke.[3] Hoping that under favorable conditions wall collisions are the only ones effecting relaxation, one would expect a relaxation time as long as the average time necessary for an atom to diffuse to the wall. This report will deal with experiments on sodium atoms which were contained in argon gas of a pressure between 1 and 40 cm Hg, while the partial sodium pressure was of the order of 10^{-7} mm Hg. Relaxation times as long as 0.21 sec were observed which in future radio-frequency resonance *reorientation* experiments should allow the observation of extraordinarily narrow lines.

The experimental arrangement was the following one. Sodium resonance radiation from a battery-operated GE sodium arc (Na 1) mounted in a Dewar was focused by a large condenser lens through a Polaroid circular polarizing sheet on a spherical absorption vessel. The volume of the thorougly baked, sealed off vessels containing some metallic sodium and the buffer varied between 0.1 and 1 liter. The vessels were generally heated to such a temperature (130–250°C) that about 50% absorption occurred. The transmitted light[3a] was focused upon a vacuum photocell whose output was amplified by a single-stage broad-band amplifier (0.05 to 1000 cps pass-band) and displayed on an oscilloscope. The earth field in the region of the absorption vessel was cancelled out and a small field of about 0.5 gauss was applied parallel to the light beam by means of a ring coil energized from a short-time-constant circuit. This arrangement now functioned in the following fashion:

As sufficient numbers of circularly polarized quanta are absorbed, the more strongly absorbing magnetic sublevels of the sodium ground state are depopulated while the less absorbing ones are filled up at their expense until a saturation polarization is attained (optical pumping).[4,5] Consequently, while this is going on, the initially transmitted light intensity I_1 will increase to a value $I_1+\Delta I$. If the axial magnetic field is now suddenly reversed, the polarization of the atoms due to its associated magnetization will follow adiabatically. This leads to a nearly instantaneous exchange of the population of the $+m$ with that of the $-m$ levels and now an overpopulation of the more strongly absorbing ones. The result is a sudden decrease of the transmitted intensity from $I_1+\Delta I$ to $I_1-\Delta I$. However, under the continued irradiation it will not take long before the original polarization pointing in the "right" direction is restored (Fig. 1). The time involved, if depolarizing relaxation effects are assumed to be insignificant, would be expected to be inversely proportional to the incident light intensity I_0. This state of affairs is found to prevail for the high-intensity oscilloscope traces. If one now reduces the light intensity further and further, there will come a time when the inverted polarization will also decay appreciably because of relaxation effects. Figure 2 shows a plot of "decay times" *versus* relative intensity. The values which were obtained from the oscilloscope traces by the construction explained in Fig. 1, converge against a characteristic relaxation time T.

A brief, more quantitative description of the processes sketched above will now be attempted. The optical pumping is appreciably modified by collisions of the excited atoms with the buffer gas. Experimental[6] evidence indicates that about 5 mm of argon is sufficient to disorient largely the orbital angular momenta with respect to the spin and also a fixed direction before reemission can occur. The situation at lower argon pressures, where only a few collisions in the excited state occur, is rather complex. However, studies of the optical pumping by investigating the polarization of the resonance fluorescence and an analysis of the collision process have recently been undertaken.[7] In the present work, a greatly simplifying assumption will be made which, even in the pressure range of interest here, 1–40 cm Hg, should hold strictly only for sufficiently strong coupling between the sodium nucleus and the electronic momenta. The assumption is that the collisions bring about a completely random redistribution of the populations of the 24 hfs magnetic sublevels of the excited ^2P state regardless of which of them had been excited originally. Under these assumptions the resonance fluorescence is completely depolarized and the rates at which the atoms return to any of the 8 hfs magnetic sublevels of the $^2S_{\frac{1}{2}}$ ground state will be identical and equal to $\frac{1}{8}$ of the sum of the probabilities for all upwards transitions. The probabilities for leaving a given ground-state m level owing to $\Delta m=+1$ transitions to the excited state under the combined influence

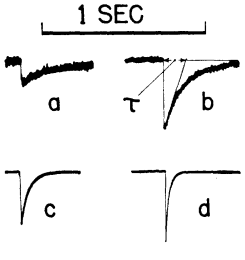

FIG. 1. Oscilloscope traces of transmitted resonance light intensity I_1 *versus* time. The time axis runs from left to right, the scale being the same for all traces. The relative intensity I_0 of the applied resonance light increased for traces a to d from 4 over 16 and 64 to 160. The sudden intensity drop occurred when the magnetic field, to which the sodium absorption cell was exposed, was instantaneously reversed. The construction shown—for trace b only—served to determine the experimental decay times τ.

[1] J. P. Wittke and R. H. Dicke, Phys. Rev. **96**, 530 (1954).
[2] H. G. Dehmelt, Phys. Rev. **99**, 527 (1955).
[3] J. P. Wittke and R. H. Dicke, Phys. Rev. **103**, 620 (1956).
[3a] Compare H. C. Dehmelt, Phys. Rev. **103**, 1125 (1956).
[4] A. Kastler, J. phys. radium **11**, 255 (1950); and Proc. Phys. Soc. (London) **A67**, 853 (1954).
[5] W. B. Hawkins, Phys. Rev. **98**, 478 (1955).
[6] W. Lochte-Holtgreven, Ann. Physik **47**, 362 (1928).
[7] Brossel, Margerie, and Kastler, Compt. rend. **241**, 865 (1955); P. L. Bender, thesis, Princeton University (1956).

of the D_1 and D_2 radiation can be computed from standard formulas[8] for the transition probabilities and are denoted by P_m.

One obtains

$$(P_{-1}:P_0:P_{-1})_{F=1}:(P_{-2}:P_{-1}:P_0:P_{+1}:P_{+2})_{F=2}$$
$$=((1+5R):(2+4R):(3+3R)):$$
$$((4+2R):(3+3R):(2+4R):(1+5R):6R),$$

R denoting the ratio of the intensities for the D_2 and D_1 components of the radiation which is expected to be close to unity.[5] The time dependence of the relative populations $a_m(t)$ of the m levels will then be described by the 8 simultaneous equations:

$$\dot{a}_m=-a_mP_m+\tfrac{1}{8}\sum a_iP_i+(1/T)(1-a_m).$$

The sum here is over the 8 sublevels of the ground state and $a_m(0)$ has been put equal to 1. The last term takes into account the relaxation processes. For the saturation distribution, $\bar{a}_m$ follows with $P_m\to\infty$ and $\dot{a}_m(\infty)=0$:

$$(\bar{a}_{-1}:\bar{a}_0:\bar{a}_{+1}):(\bar{a}_{-2}:\bar{a}_{-1}:\bar{a}_0:\bar{a}_{+1}:\bar{a}_{+2})$$
$$=(P_{-1}^{-1}:P_0^{-1}:P_{+1}^{-1}):$$
$$(P_{-2}^{-1}:P_{-1}^{-1}:P_0^{-1}:P_{+1}^{-1}:P_{+2}^{-1})$$
$$\approx((1-\bar{e}):1:(1+\bar{e})):$$
$$((1+2\bar{e}):(1+\bar{e}):1:(1-\bar{e}):(1-2\bar{e})).$$

For the last line and in the following discussion it has been assumed that

$$\bar{e}=(R-1)/(2+4R)\ll 1.$$

Instantaneous population distributions $a_m(t)$ are described in identical fashion as the saturation distribution, except that for $\bar{e}$ one has to substitute $e(t)$ which obeys the relation

$$\dot{e}=cI_0(\bar{e}-e)-(1/T)e,$$

c being a constant depending upon the P_m. The equilibrium value e_I, when the optical pumping is just compensated by relaxation, can be obtained by putting $\dot{e}=0$:

$$e_I=cI_0T\bar{e}/(cI_0T+1).$$

[8] E. U. Condon and G. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1935), Chaps. 9 and 16.

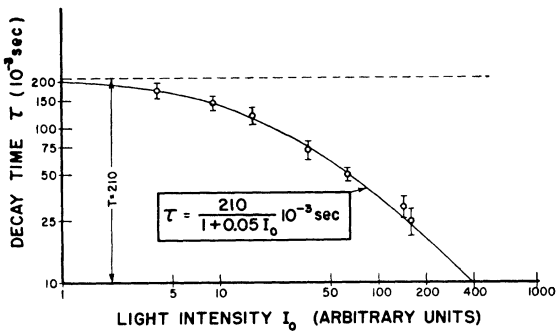

FIG. 2. Plot of the experimental decay time τ of the spin polarization against light intensity I_0. The theoretical curve has been chosen for best fit and allows the determination of the spin relaxation time T as distinct from τ, which still reflects reorientation due to impinging light quanta.

Immediately after the field reversal, we have

$$(\dot{e})_0=cI_0(\bar{e}+e_I)+(1/T)e_I;$$

from this follows that

$$2e_I/(\dot{e})_0\equiv\tau=T/(cI_0T+1),$$

which, as $\Delta I(t)$ can be shown to be proportional to $e(t)$, gives the justification for the determination of τ from the oscilloscope traces, as described in Fig. 1.

So far, quantitative measurements have been undertaken only on a 1-liter, 3-cm Hg argon sample and a 0.1-liter, 40-cm Hg argon sample. No dependence of the relaxation times on temperature, and consequently on sodium vapor pressure, was noticed in either case in the operating range. For both samples the relaxation times found were significantly shorter than the average wall diffusion times of about 0.4 sec and 0.9 sec, respectively. The relaxation times observed, namely 0.21 sec for the 3 cm Hg sample and 0.02 sec for the 40 cm Hg sample are in qualitative agreement with a relaxation mechanism due to sodium argon collisions, which would be expected to be proportional to argon pressure.

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