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Spin Exchange in a Cesium-Electron System*

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A spin-exchange optical pumping experiment to study collisions between cesium atoms and quasifree electrons is reported. In this experiment, electrons in a weak magnetic field were polarized through spin-exchange collisions with optically pumped cesium atoms. The cesium-electron collisions were the principal source of the electron-resonance linewidth, and they also gave rise to a shift in the center frequency of the electron resonance. The magnitudes of the linewidth and frequency shift depend upon the two-body scattering amplitude for elastic collisions, the cesium polarization, and the cesium density. From measurements of the electron linewidth and the frequency shift, a value is derived for the electron-cesium spin-flip cross section. The spin-flip cross section at 20°C is found to be approximately 3.5×10^{-14} cm^2. The expressions for the linewidth and frequency shift due to spin-exchange collisions of electrons with alkali-metal atoms are generalized to cover the case where there is a spin-orbit interaction between the electron–alkali-metal-atom systems.

INTRODUCTION

RECENTLY, interest in the scattering of low-energy electrons from alkali atoms has increased. This interest has resulted in the appearance in the literature of new measurements of the momentum transfer cross section for the scattering of low-energy electrons by cesium atoms[1,2] and theoretical calculations for the electron-cesium scattering phase shifts.[3,4] Values for the electron-cesium momentum transfer cross section in the temperature range of 450 to 550°K were obtained by Chen and Raether using microwave techniques to measure the electrical conductivity of a cesium-electron plasma. Flavin and Meyerand used the electron cyclotron resonance to measure the same cross section in the temperature range of 650 to 975°K. This paper reports a spin-exchange optical-pumping experiment performed to study the elastic collisions of electrons and cesium atoms at temperatures near 300°K.

In this experiment, cesium vapor in a magnetic field is polarized by the absorption of circularly polarized optical-resonance radiation incident along the direction of the magnetic field. The polarization is monitored by observing the transmission of the resonance radiation through the flask containing the cesium atoms. Free electrons produced in the flask by the ionizing radiation of tritium or by a radio-frequency discharge are polarized by spin-exchange collisions with the cesium atoms. If a radio-frequency field is adjusted so as to depolarize the electrons, the cesium atoms will also be partially depolarized through the spin-exchange collisions with the electrons, and the intensity of the light transmitted through the flask will decrease accordingly. Under suitable conditions, the electron-cesium collisions dominate other electron spin relaxation mechanisms, such as collisions with the buffer gas, and are the principal source of the width of the electron resonance signal. They also give rise to a shift in the resonance frequency. Both the frequency shift and the linewidth are functions of the electron-cesium scattering phase shifts. If the scattering is entirely *s* wave, measurements of the frequency shift and of the linewidth in the limit of zero radio-frequency field yield a value for the spin-flip cross section even if the number of cesium atoms in the flask is unknown.

In a recent paper a complete theoretical treatment of spin-exchange optical-pumping experiments was given, and an experimental study of rubidium-electron collisions was reported.[5] The experiments reported in the present paper are similar to the experiments reported in that paper. The agreement of the cesium-electron measurements with the theory is not completely satisfactory, however. One possible source of this discrepancy could be a spin-orbit interaction in the electron-cesium system. The previous theory assumed there was no spin-orbit interaction. In this paper the theoretical expressions for the electron linewidth and frequency shift are generalized to cover the situation where there is spin-orbit coupling. The first part of this paper summarizes and extends the theory; the second part summarizes the measurements; the third part compares the results of these measurements on the electron-cesium system with the available theoretical calculations and other measurements.

THEORY

In this section we will be primarily concerned with summarizing the theoretical expressions for the electron linewidth and frequency shift and generalizing them to the case where there is spin-orbit coupling. We shall assume that the alkali metal or cesium atom has no nuclear spin. The basic problem in this calculation is to determine the rate of change of the cesium and electron spin-space density matrices due to spin-

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[2] R. K. Flavin and R. G. Meyerand, Jr., IEEE Thermionic Converter Specialist Conference, Gatlinburg, Tennessee, October 1963 (unpublished).

[3] L. B. Robinson, Phys. Rev. **127**, 2076 (1962).

[4] P. M. Stone and J. R. Reitz, Phys. Rev. **131**, 2101 (1963).

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exchange collisions. For a more complete treatment of the theory the reader should refer to the previous paper.[5]

To describe the electron-cesium scattering, it is convenient to employ the center-of-mass system and to use the coordinates of the electron relative to the cesium atom. The incoming wave for the electron-cesium scattering problem can be written in the form

$$\frac{1}{L^{3/2}}\exp(i\mathbf{k}_0\cdot\mathbf{r})|s_0\rangle, \tag{1}$$

where s_0 is the initial electron-cesium atom spin state and the normalization is such that we have one electron in the box. The scattered wave will be

$$\psi(r,\mathbf{k},s) = \frac{1}{L^{3/2}}\left[\exp(i\mathbf{k}_0\cdot\mathbf{r})|s_0\rangle + \left(\frac{e^{ikr}}{r}\right)M_{ss_0}(\mathbf{k};\,\mathbf{k}_0)|s_0\rangle\right], \tag{2}$$

where $M_{ss_0}(\mathbf{k};\,\mathbf{k}_0)$ is a function of the angle between $\mathbf{k}$ and $\mathbf{k}_0$ and is in general a matrix in spin space which allows for the possibility of changes in spin states during a collision. In the earlier paper it was shown that in terms of the M matrix the time rate of change of the electron density matrix due to spin-exchange collisions with the alkali-metal atom A was

$$\begin{aligned} d\rho(e)/dt = v_{eA}N_A\,\mathrm{Tr}_A\Big[& (2\pi i/k)[M(\theta=0,\,\phi=0)\rho(e,A) \\ & -\rho(e,A)M^\dagger(\theta=0,\,\phi=0)] \\ & + \int d\Omega M(\theta,\phi)\rho(e,A)M^\dagger(\theta,\phi)\Big]. \end{aligned} \tag{3}$$

Here v_{eA} is the relative velocity of the electrons and the alkali-metal atoms, N_A is the number of alkali atoms per cm^3, Tr_A stands for the trace over the alkali-atom spin coordinates, $\rho(e,A)$ is the electron-alkali atom spin-space density matrix, $M(\theta,\phi)$ is an abbreviation for $M_{ss_0}(k;\,k_0)$, and θ, ϕ are the polar coordinates of the vector $\mathbf{k}$ with respect to $\mathbf{k}_0$. An analogous expression for the time-rate change of the alkali-metal atom can be derived by interchanging A and e. In the earlier paper it was assumed that there was no spin-orbit coupling and that M could be written in the form

$$M = f_3(\theta)P_3 + f_1(\theta)P_1, \tag{4}$$

where P_3 and P_1 are the projection operators for the triplet and singlet electronic states, f_3 and f_1 are the triplet and singlet scattering amplitudes. In terms of the Pauli spin matrices the projection operators are

$$P_3 = \tfrac{1}{4}(3+\boldsymbol{\sigma}_e\cdot\boldsymbol{\sigma}_A), \tag{5}$$

and

$$P_1 = \tfrac{1}{4}(1-\boldsymbol{\sigma}_e\cdot\boldsymbol{\sigma}_A). \tag{6}$$

In terms of the triplet and singlet phase shifts, the scattering amplitudes are

$$f_3 = (1/2ik)\sum_{l=0}^{\infty}(2l+1)(e^{2i\delta_l^3}-1)P_l(\cos\theta), \tag{7}$$

and

$$f_1 = (1/2ik)\sum_{l=0}^{\infty}(2l+1)(e^{2i\delta_l^1}-1)P_l(\cos\theta). \tag{8}$$

It was further shown that if the alkali metal atom was replaced by an equivalent spin-$\frac{1}{2}$ system, the time dependence of the electron density matrix was given by the expression

$$\frac{d\rho(e)}{dt} = \begin{bmatrix} \dfrac{P(A)-P(e)}{2T_{ee}} & -\dfrac{1-i\kappa P(A)}{T_{ee}}\rho_{12}(e) \\ -\dfrac{1+i\kappa P(A)}{T_{ee}}\rho_{21}(e) & \dfrac{P(e)-P(A)}{2T_{ee}} \end{bmatrix}, \tag{9}$$

where $P(A)$ and $P(e)$ are the electron polarization and the alkali-atom electronic polarization given by the equations

$$P(e) = \rho_{11}(e) - \rho_{22}(e), \tag{10}$$

and

$$P(A) = \rho_{11}(A) - \rho_{22}(A), \tag{11}$$

and the spin-exchange relaxation time is given by the equation

$$1/T_{ee} = v_{eA}N_A\sigma_{\mathrm{SF}}. \tag{12}$$

In terms of the phase shifts, the spin-flip cross section σ_{SF} and the frequency shift parameter κ are given by the equations

$$\sigma_{\mathrm{SF}} = (\pi/k^2)\sum_{l=0}^{\infty}(2l+1)\sin^2(\delta_l^3-\delta_l^1), \tag{13}$$

and

$$\kappa = (1/\sigma_{\mathrm{SF}})(\pi/2k^2)\sum_{l=0}^{\infty}(2l+1)\sin 2(\delta_l^3-\delta_l^1). \tag{14}$$

The corresponding equations for the time rate of change of the alkali-atom density matrix are

$$\frac{d\rho(A)}{dt} = \begin{bmatrix} \dfrac{P(e)-P(A)}{2T_{eA}} & 0 \\ 0 & \dfrac{P(A)-P(e)}{2T_{eA}} \end{bmatrix}, \tag{15}$$

where

$$1/T_{eA} = v_{eA}N_e\sigma_{\mathrm{SF}}. \tag{16}$$

In this paper we wish to derive the generalization of these expressions when there is also spin-orbit coupling. We shall assume that the potential which describes the interaction between the electron and the alkali atom can be written in the form

$$V(r)+V_1(r)\mathbf{S}\cdot\mathbf{L}, \tag{17}$$

where $\mathbf{L}$ is the orbital angular momentum of the electron with respect to the rubidium atom and

$$\mathbf{S}=\tfrac{1}{2}(\boldsymbol{\sigma}_e+\boldsymbol{\sigma}_A). \tag{18}$$

For an interaction of this form there will be a different phase shift for each of the total angular-momentum states derived by coupling $\mathbf{S}$ to $\mathbf{L}$. If we introduce the total angular momentum,

$$\mathbf{J}=\mathbf{S}+\mathbf{L}, \tag{19}$$

then in terms of the notation

$$P_{l,j}{}^{2s+1}, \tag{20}$$

the projection operators for the states of total angular momentum j derived from a total spin state s and an angular-momentum state l by coupling $\mathbf{S}$ and $\mathbf{L}$ together in various fashions can be written in the form

$$P_{l,l}{}^{1}=1, \tag{21}$$

$$P_{l,l+1}{}^{3}=\frac{(\mathbf{S}\cdot\mathbf{L}+1)(\mathbf{S}\cdot\mathbf{L}+l+1)}{(l+1)(2l+1)}, \tag{22}$$

$$P_{l,l}{}^{3}=\frac{(l-\mathbf{S}\cdot\mathbf{L})(\mathbf{S}\cdot\mathbf{L}+l+1)}{l(l+1)}, \tag{23}$$

$$P_{l,l-1}{}^{3}=\frac{(\mathbf{S}\cdot\mathbf{L}-l)(\mathbf{S}\cdot\mathbf{L}+1)}{l(2l+1)}. \tag{24}$$

In terms of these projection operators the M matrix is

$$M(\theta,\phi)=(1/k)\{[\sum_{l=0}^{\infty}(2l+1)e^{i\delta_{ll}{}^1}\sin\delta_{ll}{}^1P_l(\cos\theta)]\tfrac{1}{4}(1-\boldsymbol{\sigma}_e\cdot\boldsymbol{\sigma}_A)+[e^{i\delta_{01}{}^3}\sin\delta_{01}{}^3+\sum_{l=1}^{\infty}(2l+1)(e^{i\delta_{l,l+1}{}^3}\sin\delta_{l,l+1}{}^3P_{l,l+1}{}^3$$

$$+e^{i\delta_{l,l}{}^3}\sin\delta_{l,l}{}^3P_{l,l}{}^3+e^{i\delta_{l,l-1}{}^3}\sin\delta_{l,l-1}{}^3P_{l,l-1}{}^3)P_l(\cos\theta)]\tfrac{1}{4}(3+\boldsymbol{\sigma}_e\cdot\boldsymbol{\sigma}_A)\}. \tag{25}$$

If this expression is inserted into Eq. (3) and we retain only states of orbital angular momentum 0 and 1, the following expression is obtained for the time rate of change of the electron density matrix:

$$\frac{d\rho(e)}{dt}=\begin{bmatrix} \dfrac{P(A)-P(e)}{2T_{ee1}}-\dfrac{P(A)+P(e)}{2T_{ee3}} & -\dfrac{1-i\kappa_1P(A)}{T_{ee2}}\rho_{12}(e) \\ -\dfrac{1+i\kappa_1P(A)}{T_{ee2}}\rho_{21}(e) & \dfrac{P(e)-P(A)}{2T_{ee1}}+\dfrac{P(A)+P(e)}{2T_{ee3}} \end{bmatrix}, \tag{26}$$

where

$$1/T_{ee1}=v_{eA}N_A(\pi/k^2)[\sin^2(\delta_{01}{}^3-\delta_{00}{}^1)+2\sin^2(\delta_{12}{}^3-\delta_{11}{}^1)+\sin^2(\delta_{10}{}^3-\delta_{11}{}^1)], \tag{27}$$

$$1/T_{ee3}=v_{eA}N_A(\pi/k^2)[3\sin^2(\delta_{12}{}^3-\delta_{11}{}^3)], \tag{28}$$

$$1/T_{ee2}=v_{eA}N_A(\pi/k^2)[\sin^2(\delta_{01}{}^3-\delta_{00}{}^1)+\tfrac{3}{2}[\sin^2(\delta_{12}{}^3-\delta_{11}{}^1)+\sin^2(\delta_{11}{}^3-\delta_{11}{}^1)]+\sin^2(\delta_{11}{}^3-\delta_{10}{}^3)+\tfrac{1}{2}\sin^2(\delta_{12}{}^3-\delta_{11}{}^3)], \tag{29}$$

and

$$\frac{\kappa_1}{T_{ee2}}=v_{eA}N_A(\pi/2k^2)[\sin 2(\delta_{01}{}^3-\delta_{00}{}^1)+\tfrac{3}{2}[\sin 2(\delta_{12}{}^3-\delta_{11}{}^1)+\sin 2(\delta_{11}{}^3-\delta_{11}{}^1)]$$

$$+\sin 2(\delta_{12}{}^3-\delta_{10}{}^3)+\tfrac{3}{2}\sin 2(\delta_{11}{}^3-\delta_{12}{}^3)]. \tag{30}$$

These expressions are considerably more complicated than the corresponding expressions [Eq. (9)] when there is no spin-orbit coupling. The relaxation time for the off-diagonal elements of the electron density matrix is no longer simply related to the relaxation time for the diagonal elements. The expression for the ratio of the frequency shifts to the linewidth is more complicated. The corresponding expression for the time rate of change of the alkali-atom density matrix is

$$\frac{d\rho(A)}{dt}=\begin{bmatrix} \dfrac{P(e)-P(A)}{2T_{eA1}}-\dfrac{P(A)+P(e)}{2T_{eA3}} & 0 \\ 0 & \dfrac{P(A)-P(e)}{2T_{eA1}}+\dfrac{P(A)+P(e)}{2T_{eA3}} \end{bmatrix}. \tag{31}$$

Here

$$1/T_{eA1}=(N_e/N_A)(1/T_{ee1}),\quad\text{and}\quad 1/T_{eA3}=(N_e/N_A)(1/T_{ee3}).$$

If we now use the phenomenological equations

$$\frac{d\rho(e)}{dt}=\begin{bmatrix} \dfrac{\frac{1}{2}-\rho_{11}(e)}{T_{1e}} & -\dfrac{\rho_{12}(e)}{T_{2e}} \\ -\dfrac{\rho_{21}(e)}{T_{2e}} & \dfrac{\frac{1}{2}-\rho_{22}(e)}{T_{1e}} \end{bmatrix} \tag{32}$$

and

$$\frac{d\rho(A)}{dt}=\begin{bmatrix} \dfrac{\frac{1}{2}-\rho_{11}(A)}{T_{1A}} & -\dfrac{\rho_{12}(A)}{T_{2A}} \\ -\dfrac{\rho_{21}(A)}{T_{2A}} & \dfrac{\frac{1}{2}-\rho_{22}(A)}{T_{1A}} \end{bmatrix} \tag{33}$$

to represent the other relaxation mechanisms, we can use the procedure of the previous paper to derive an expression for the change in light transmitted by the absorption flask when there is a radio-frequency field which can depolarize the electrons. If we assume that there is a static magnetic field H_0 along the direction of the light beam, and a radio-frequency field $2H_1 \cos\omega t$ perpendicular to the light beam, the change in transmitted light is

$$\begin{aligned}\delta I_T=AA_\alpha(s)\left(\int_0^\infty I(\nu,0)d\nu\right)&\left[\frac{\tau^{-1}}{\tau^{-1}+T_{eA1}^{-1}+T_{eA3}^{-1}+T_{1A}^{-1}}\right]\\ &\times\left[\frac{T_{eA1}^{-1}-T_{eA3}^{-1}}{T_{1A}^{-1}+T_{eA1}^{-1}+T_{eA3}^{-1}-(T_{eA1}^{-1}-T_{eA3}^{-1})((T_{ee1}^{-1}-T_{ee3}^{-1})/(T_{ee1}^{-1}+T_{ee3}^{-1}+T_{1e}^{-1}))}\right]\\ &\times\left[\frac{T_{ee1}^{-1}-T_{ee3}^{-1}}{T_{ee1}^{-1}+T_{ee3}^{-1}+T_{1e}^{-1}}\right]\left[\frac{\omega_1^2\tau_1\tau_2}{1+\omega_1^2\tau_1\tau_2+(\tau_2)^2(\omega_0-\delta\omega_0-\omega)^2}\right].\end{aligned} \tag{34}$$

Here

$$\tau_1=\frac{\tau^{-1}+T_{1A}^{-1}+T_{eA1}^{-1}+T_{eA3}^{-1}}{(\tau^{-1}+T_{1A}^{-1}+T_{eA1}^{-1}+T_{eA3}^{-1})(T_{1e}^{-1}+T_{ee1}^{-1}+T_{ee3}^{-1})-(T_{eA1}^{-1}-T_{eA3}^{-1})(T_{ee1}^{-1}-T_{ee3}^{-1})}, \tag{35}$$

and

$$\tau_2^{-1}=T_{2e}^{-1}+T_{ee2}^{-1}. \tag{36}$$

In these equations τ is the pumping time, A is the cross-sectional area of the cylindrical absorption flask, $A_\alpha(s)$ is a function which gives the fraction of the incident light absorbed by the flask, $I(\nu,0)$ is the intensity per unit frequency range of the circularly polarized D_1 light incident on the absorption flask, ω_1 is the resonance frequency of the electrons in the field H_1, ω_0 is the resonance frequency of the electrons in the field H_0, and $\delta\omega_0$ is the frequency shift due to spin exchange collisions.[6] In terms of the phase shifts

$$\delta\omega_0=P(A)(\kappa_1/T_{ee2}). \tag{37}$$

EXPERIMENTAL PROCEDURE AND RESULTS

The apparatus used in this experiment is described in the previous paper on the rubidium-electron system. The experiments were carried out in a magnetic field of 50 mG. The magnetic field was sufficiently homogeneous that the full width at half-maximum of the cesium Zeeman transitions (350 cps/mG) was 40 cps. The resonance signal was measured by amplitude modulating the radio-frequency field with a mercury relay and observing the demodulated absorption signal with a lock-in detector. The temperatures of the absorption flasks were measured with a copper-constantan thermocouple which was attached to the side of the bulb. Solid carbon dioxide was employed to reach temperatures below 20°C.

Two types of absorption bulbs were used. The first type was a 500-cm³ spherical flask containing cesium, 41±1 mm Hg of a helium buffer gas, and 2 C of tritium. The free electrons were produced through ionization by the tritium beta rays. The second type of absorption bulb consisted of a 300-cm³ flask which was connected by a neck 1 cm in diameter and 1 cm long to a 25-cm³ bulb. The 25-cm³ bulb was constructed with two glass-covered tungsten electrodes, and contained 40.6 mm Hg of a helium buffer gas. A continuous radio-frequency discharge between these two electrodes provided a source of free electrons. The electrons then diffused through the connecting neck into the main absorption flask. This second type of flask was constructed primarily to ensure that the measurements were made on

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[6] The corresponding equations in Ref. 5 [Eqs. (77) and (79)] contain a misprint. The expression $T_{ee}/(T_{ee}+T_{1R})$ should read $T_{ee}/(T_{ee}+T_{1e})$.

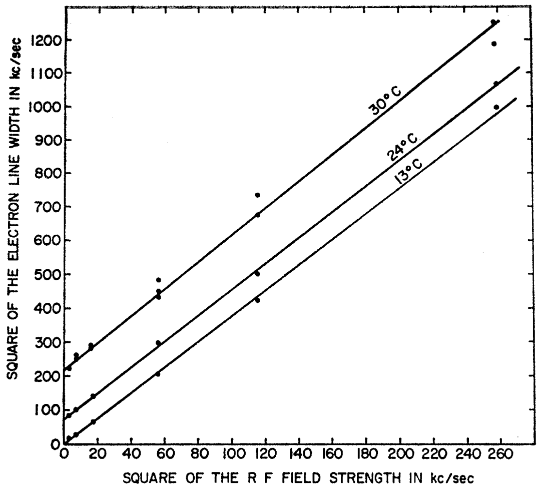

FIG. 1. The square of the full width at half-maximum of the electron line in kc/sec plotted versus the square of the rf field strength in kc/sec for three different temperatures. The slope of these lines is equal to $4\tau_1/\tau_2$; the intercept of the lines is $(1/\pi\tau_2)^2$. The measurements were made in a tritium-helium bulb.

thermal electrons. Identical results were obtained with both types of bulbs. Since there was less background light with the tritium bulbs, they were used for most of the measurements.

The first measurements were directed toward a determination of τ_1 and τ_2 as a function of temperature. According to Eq. (34) the amplitude of the electron resonance signal is described by the equation

$$\delta I_T = (\text{const})\frac{\omega_1^2\tau_1\tau_2}{1+\omega_1^2\tau_1\tau_2+(\omega_0-\delta\omega_0-\omega)^2(\tau_2)^2}. \tag{38}$$

This equation indicates a method for determining τ_1 and τ_2. A plot of the square of the full width at half-maximum of the electron line versus $(\omega_1/2\pi)^2$ should yield a straight line with a slope of $4\tau_1/\tau_2$ and with a zero radio-frequency intercept of $(1/\pi^2\tau_2^2)$. In order to determine these two relaxation times, measurements were made of the electron linewidth as a function of the strength of the radio-frequency field. Runs were made at various temperatures using both the tritium and the discharge bulbs. The radio-frequency field strength ω_1 was obtained by keeping the radio-frequency oscillator set at the electron frequency and increasing the magnetic field H_0 until the cesium signal was visible on the oscilloscope. The cesium signal was then photographed. This signal showed the characteristic modulation of the pumping light due to the nutation of the cesium moment. Since the magnetic moment of the electron is approximately eight times the atomic moment of the cesium atom (nuclear spin equals $\frac{7}{2}$), ω_1 was obtained from the relationship

$$\omega_1 = 16\pi\nu(\text{cesium}). \tag{39}$$

Figure 1 shows several plots of (linewidth)^2 versus $(\omega_1/2\pi)^2$ for various bulb temperatures. The slopes of the lines indicate that

$$\tau_1/\tau_2 = 1.0 \pm 0.1$$

for all the measured temperatures. Table I summarizes the values of τ_2 determined from the zero radio-frequency field intercepts.

TABLE I. The values of τ_2 obtained for various bulb temperatures. The values of τ_2 referred to 20°C were obtained from the equation $\tau_2(20°)/\tau_2(T) = N_{\text{Cs}}(T)/N_{\text{Cs}}(20°)$. Here $N_{\text{Cs}}(T)$ is the number of cesium atoms per cm^3 at temperature T.

Bulb type	Temperature in °C	τ_2 in sec	τ_2 (20°C) in sec
Tritium-helium	12.5	14.2×10^{-5}	6.5×10^{-5}
Tritium-helium	16	9.6×10^{-5}	6.3×10^{-5}
Tritium-helium	20	5.8×10^{-5}	5.8×10^{-5}
Tritium-helium	24	3.74×10^{-5}	5.5×10^{-5}
Tritium-helium	30	2.15×10^{-5}	5.8×10^{-5}
Helium -discharge	30	2.15×10^{-5}	5.8×10^{-5}

Figure 2 shows a plot of $1/\pi\tau_2$ as a function of temperature. According to Eqs. (36) and (29)

$$1/\tau_2 = 1/T_{2e} + 1/T_{ee2},$$

and

$$\frac{1}{T_{ee2}} \alpha\, N_A v_{eA}\frac{1}{k^2} \alpha \frac{p(\text{Cs})}{T^{3/2}}, \tag{40}$$

where $p(\text{Cs})$ is the cesium vapor pressure. The dashed curve is a plot of $p(\text{Cs})T^{-3/2}$ versus temperature and it has been normalized to fit the data in the middle of the temperature range. Table I shows in another fashion the agreement between the measured temperature dedependence of the linewidth and the temperature dependence expected if the linewidth is due entirely to spin-exchange collisions. We conclude that $\tau_2 = T_{ee2}$ in

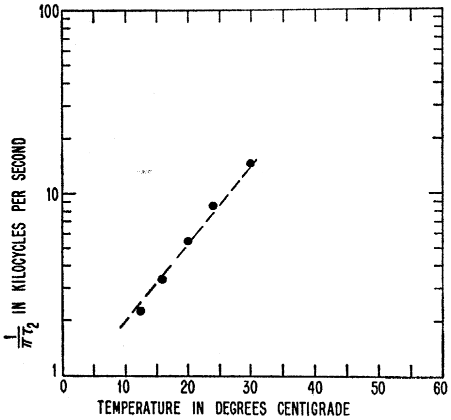

FIG. 2. A plot of $1/\pi\tau_2$ as a function of temperature. The values of $1/\pi\tau_2$ were obtained at each temperature by measuring the zero rf field intercepts of straight line plots such as those shown in Fig. 1. The dashed curve represents the temperature dependence of the function, $p(\text{cesium})T^{-3/2}$. This is the temperature dependence one expects for $1/\pi\tau_2$ if spin-exchange collisions are the principal source of the linewidth.

TABLE II. The measured values of the frequency shift $\delta\nu_0$ at various temperatures. Also listed are the corresponding values of $P(\mathrm{Cs})\kappa$ obtained from the relation $P(\mathrm{Cs})\kappa_1=2\pi T_{ee1}\delta\nu_0=2\delta\nu_0/\Delta\nu$. Here $P(\mathrm{Cs})$ is the electronic polarization of the cesium and $\Delta\nu$ is the full width at half-maximum of the electron resonance.

Bulb type	Temperature in °C	$\delta\nu_0$ (cps)	$P(\mathrm{Cs})\kappa_1$
Tritium-helium	11	10	−0.011
Tritium-helium	17	23	−0.012
Tritium-helium	20	55	−0.021
Tritium-helium	24.5	65	−0.014
Discharge-helium	30	190	−0.025

the temperature range investigated and that the electron relaxation is dominated by spin-exchange collisions.

The frequency shift due to electron-cesium collisions was measured by first observing the electron resonance frequency with left circularly polarized light and then with right circularly polarized light. This operation changed the sign of the cesium polarization and consequently the direction of the frequency shift. The shift was determined from the equation

$$\delta\nu_0=\frac{\nu(\mathrm{left})-\nu(\mathrm{right})}{2}. \qquad (41)$$

During measurements of the frequency shift, measurements of the cesium Zeeman frequency were interspersed to correct for drifting of the magnetic field. The electron resonance frequency was higher when left circularly polarized light was incident on the absorption flask and the cesium polarization was positive. Table II summarizes the measured values of the frequency shift. Table II also lists the value of $P(\mathrm{Cs})\kappa_1$ computed from the measured linewidths and frequency shifts by using the equation

$$P(\mathrm{Cs})\kappa_1=2\pi T_{ee2}(\delta\nu_0)=2\delta\nu_0/\Delta\nu, \qquad (42)$$

where $\Delta\nu$ is the full width at half-maximum of the electron line. The computed values indicate that $2\delta\nu_0/\Delta\nu$ varies slowly with temperature. Figure 3 shows a plot of the measured frequency shifts versus the bulb temperature. Figure 3 also shows the expected temperature dependence if the frequency shift depends only on spin-exchange collisions with the cesium atoms.

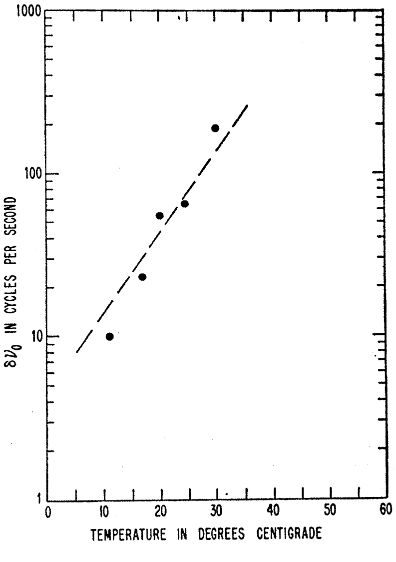

FIG. 3. A plot of the frequency shift $\delta\nu_0$ as a function of temperature. The dashed line represents the observed temperature dependence of $(1/\pi\tau_2)$. One expects the shift to have this same temperature dependence.

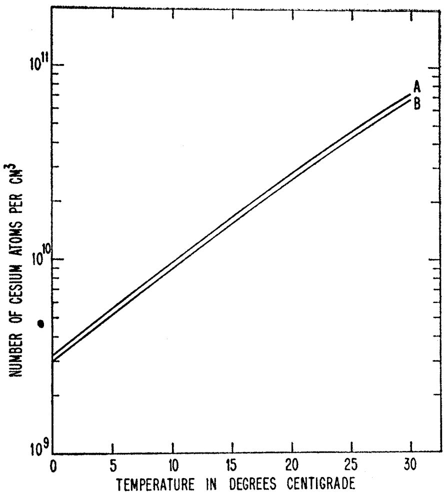

FIG. 4. Plots of the cesium density as a function of temperature. Curve A is derived from the equation (Ref. 7)

$$\log_{10}p(\mathrm{Cs})=(-4041/T)+11.053-1.35\log_{10}T,$$

where $p(\mathrm{Cs})$ is the vapor pressure of cesium in mm Hg and T is the temperature in degrees Kelvin. Curve B is derived from the equation (Ref. 8)

$$\log_{10}p(\mathrm{Cs})=(-4075/T)+11.38-1.45\log_{10}T.$$

Measurements of the relative amplitudes of the cesium Zeeman transitions in a magnetic field sufficiently strong to separate them indicated that the cesium electronic polarization at 20°C was greater than 0.25. This measurement is subject to a systematic error since it is based on a measurement of relative signal amplitudes and not of the absolute signal strength. It is felt that the value 0.25 is a lower limit.

INTERPRETATION

In this section we wish to see what the measurements indicate concerning the cesium electron spin-flip cross section. First let us assume that the scattering is predominantly s wave and that there is no spin-orbit coupling. In this case we can use Eqs. (13) and (14) and the values of the ratio of frequency shift to the linewidth listed in Table II to determine $(\delta_0^3-\delta_0^1)$. At 20°C we find that

$$\cot(\delta_0^3-\delta_0^1)=\frac{0.02\pm0.01}{P(\mathrm{Cs})}.$$

If we assume that $P(\mathrm{Cs})\geq0.25$, then

$$-0.01\geq\cot(\delta_0^3-\delta_0^1)\geq-0.12,$$

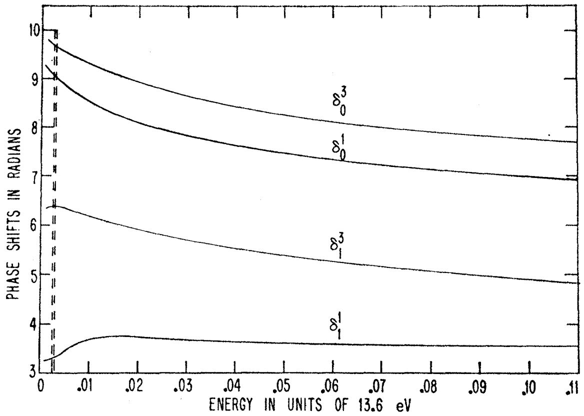

FIG. 5. The singlet and triplet phase shifts for *s*- and *p*-wave cesium-electron scattering as calculated by Stone and Reitz (Ref. 4) are plotted as a function of energy in units of 13.6 eV. The dashed lines indicate the energy range of the experiments reported in the present paper, i.e., from 0.0376 to 0.0394 eV.

and it follows that

$$(\delta_0^3-\delta_0^1)=-\tfrac{1}{2}\pi+\epsilon \quad\text{or}\quad \tfrac{1}{2}\pi+\epsilon,$$

where ϵ is positive and small.

We can obtain an alternate value for the phase shifts by using the measured values of the linewidth and an expression for the density of cesium atoms in the bulb. There are several expressions for the cesium vapor pressure in the literature; two of these expressions which are considered to be most reliable are plotted in Fig. 4.[7,8] In order to relate the observed linewidth to the theoretical expression, we must average the signal calculated assuming one velocity for the electrons over the electron velocity distribution. The observed signal is a superposition of Lorenzian lines whose widths depend upon the electron velocities. In order to perform this average, we will assume that the electrons have a Maxwell-Boltzmann velocity distribution characterized by the bulb temperature T and that the scattering phase shifts vary slowly with electron temperature. This averaging process yields an expression for the observed linewidth $\Delta\nu$ which is coincidentally the same as that obtained by averaging over a Maxwell-Boltzmann distribution the expression for the linewidth obtained by assuming one velocity for the electrons.

$$\langle\Delta\nu\rangle_{\mathrm{av}}=\left\langle\frac{1}{\pi T_{ee}}\right\rangle_{\mathrm{av}}=N_A\left(\frac{\hbar}{m}\right)^2\left(\frac{2m}{\pi kT}\right)^{1/2}\times\sum_{l=0}^{\infty}(2l+1)\sin^2(\delta_l^3-\delta_l^1)\,. \tag{43}$$

If we use the observed value of T_{ee} at 20°C and the rubidium density derived from curve A in Fig. 4, we find that

$$\sum_{l=0}^{\infty}(2l+1)\sin^2(\delta_l^3-\delta_l^1)=1.25\pm0.1\,, \tag{44}$$

where the error reflects the uncertainty in the measured linewidths. This result indicates that *s*-wave scattering alone can not account for the observed spin-flip cross section and that other angular momenta must be present. If we assume there is only *s*- and *p*-wave scattering, we should look for simultaneous solutions of the two equations

$$\sin^2(\delta_0^3-\delta_0^1)+3\sin^2(\delta_1^3-\delta_1^1)=1.25\pm0.1\,, \tag{45}$$

and

$$\frac{1}{2}\,\frac{\sin2(\delta_0^3-\delta_0^1)+3\sin2(\delta_1^3-\delta_1^1)}{\sin^2(\delta_0^3-\delta_0^1)+3\sin^2(\delta_1^3-\delta_1^1)}=-\frac{0.02\pm0.01}{P(\mathrm{Cs})} \tag{46}$$

in order to determine both the *s*- and *p*-wave contributions to the scattering. If the cesium polarization is ≥ 0.25, then there are no simultaneous solutions of Eqs. (45) and (46). The observed frequency shift is too small to allow a sufficiently large *p*-wave contribution to explain the magnitude of the observed spin-flip cross section.

If one believes the vapor-pressure curve, the spin-flip cross section at 20°C is $(3.95\pm0.32)\times10^{-14}$ cm^2. If one believes the frequency-shift measurements and the theory, one concludes that the scattering is entirely *s* wave, that the spin-flip cross section is $(3.16\pm0.25)\times10^{-14}$ cm^2 and that $|\delta_0^3-\delta_0^1|\simeq\frac{1}{2}\pi$.

There are at least two ways in which one can rationalize this failure to yield a unique solution. The first is to assume that the density of cesium atoms in the absorption flask is 25% greater than the values calculated from the measured vapor pressure curves. It is difficult to say how reliably one can determine the density of cesium atoms at such low temperatures. Another way is to assume that the spin-exchange theory is oversimplified. As an improvement in this direction we can use the theory which includes the spin-orbit coupling. A short study of the expressions for the relaxation times and frequency shift when there are *s* waves, *p* waves, and spin-orbit coupling shows that we now have a sufficient number of parameters to fit all the observations and explain a large spin-flip cross section and a small frequency shift. It would require a detailed calculation to say whether or not these values of the phase shifts are reasonable.

In Fig. 5 we have plotted, as a function of energy, the calculated values of the *s*- and *p*-wave phase shifts obtained by Stone and Reitz for elastic collisions between electrons and cesium atoms. The differences of the triplet and singlet phase shifts which they have calculated are too small to explain our results. They

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[7] G. G. Grau and K. L. Schaefer, *Landolt-Börnstein Zahlenwerte und Funktionen Aus Physik, Chemie, Astronomie, Geophysik, und Technik* (Springer-Verlag, Berlin, 1960), Vol. II, p. 7.

[8] *Metals Reference Handbook*, edited by Colin J. Smithells (Butterworths Scientific Publications Ltd., London, 1955), Vol. II, p. 613.

predict a spin-flip cross section of 1.58×10^{-14} cm² and a value for the shift parameter κ of -0.2.

Chen and Raether find that their data for the momentum-transfer cross section in the temperature range of 450 to 550°K can be fitted by the equation

$$Q_m=\frac{1.61\times10^{-10}}{T}-\frac{9.63\times10^{-12}}{T^{1/2}}+2.03\times10^{-13}\text{ cm}^2. \tag{47}$$

If one extrapolates this function to 20°C, one finds that $Q_m=1.9\times10^{-13}$ cm². Q_m is by definition related to the ordinary differential cross section $I(\theta)$ for elastic scattering by the equation

$$Q_m=\int d\Omega(1-\cos\theta)I(\theta)\,, \tag{48}$$

$$I(\theta)=\tfrac{3}{4}|f_3|^2+\tfrac{1}{4}|f_1|^2. \tag{49}$$

If one assumes that the scattering at room temperature is exclusively s wave, then one obtains

$$\begin{aligned}Q_m=\sigma_{\text{tot}}&=(\pi/k^2)[3\sin^2\delta_0{}^3+\sin^2\delta_0{}^1]\\&=3.16\times10^{-14}[3\sin^2\delta_0{}^3+\sin^2\delta_0{}^1].\end{aligned} \tag{50}$$

In any situation, however,

$$3\sin^2\delta_0{}^3+\sin^2\delta_0{}^1\leq4\,. \tag{51}$$

Hence one would conclude that there is a large p-wave contribution present. The momentum-transfer cross sections measured by Flavin and Meyerand do not overlap the data of Chen and Raether. The results of Flavin and Meyerand indicate that the momentum-transfer cross section decreases with decreasing temperature; the measurements of Chen and Raether show an increase of the cross section with decreasing temperature. There may be a sharp bend in the cross section's dependence on the temperature; in either case, however, one is reluctant to trust an extrapolation of Chen and Raether's data down to 20°C.

CONCLUSIONS

This experiment indicates that there is still some difficulty in understanding the magnitude of the electron–alkali-metal-atom spin-flip cross sections and of the electron frequency shifts due to spin-exchange collisions. It is not clear whether there is some incompleteness in the theory of the frequency shift or whether some of the measurements are in error. For both rubidium and cesium the spin-flip cross sections are large and the frequency shifts are small. In both rubidium and cesium the s-wave part of the scattering seems to be near the unitary limit and

$$|\delta_0{}^3-\delta_0{}^1|\simeq\pi/2\,.$$

It is not easy to understand this relationship. It is in general difficult to calculate the spin-flip cross section. Most of the scattering depends upon the polarization potential which is the same in both singlet and triplet states. The spin-flip cross section, however, depends upon the difference of the phase shifts and thus possibly upon the difference of two large numbers. It is interesting that the frequency shifts for electron-cesium collisions have the opposite sign from those for electron-rubidium collisions.

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