$$\frac{\mathbf{v}^2}{c^2} \ll \frac{\sinh s\xi}{s\xi} \frac{Y \exp(-X \cosh \xi)}{2X \sinh \xi + Y} \left[ I_s(X) + \xi \tanh \xi X I'_s(X) \right] \ll 1.$$
 (15)

The dispersion relation (14) becomes much simpler in the limiting case  $X \ll 1$ . We note that in spite of the assumed smallness of X we obtain a result that differs significantly from the conclusions of Korneev and Starostin.<sup>[5]</sup> The reason is that we shall use the asymptotic value of the function  $I_s(X)$ , and not the expansion of  $\exp(-X \cosh \xi)$ . Since

 $I_*(X) \approx X^*/2^* s! \qquad (X \ll 1).$ 

it follows that, putting  $\sinh \xi \approx \cosh \xi \approx (1/2)e^{\xi}$  we get

$$\Delta_s = \frac{Y(1+s\xi)}{2^{s-1}s!\,s\xi}\frac{Z^s e^{-Z^{s}2}}{Z+Y}, \quad Z = Xe^{\xi}.$$
 (16)

The maximum of this expression occurs at

$$Z = Z_s = X_s e^{\frac{1}{2}} = [\frac{1}{4}(Y + 2 - 2s)^2 + 2sY]^{\frac{1}{2}} - \frac{1}{2}(Y + 2 - 2s).$$
(17)

The condition  $X_s \ll 1$  is easily satisfied. In the limiting case  $Y \ll 1$ , the condition  $X_s \ll 1$  is automatically satisfied for the harmonic s = 1, and at s > 1 it reduces to the requirement  $e^t \gg 2(s-1)$ .

We write now the maximum value of  $\Delta_s$  at  $Y \ll 1$ :

$$(\Delta_s)_{\max} = \frac{Y}{4s_{\xi}^2} e^{-(s-1)} (1+s_{\xi}^2) \frac{(s-1)^{s-1}}{s!}.$$
 (18)

As s = 1, the last factor must be replaced by unity. In<sup>[5]</sup>, this maximum could not be obtained, since the authors confined themselves there to excessively small values of X. We therefore compare our results with the classical result obtained by Akhiezer *et al.*<sup>[11]</sup> The qualitative differences are the following: a)  $(\Delta_s)_{\max}$  decreases exponentially with increasing number of the harmonic. b) The value of  $Z_s$  increases linearly with increasing number of the harmonic. c)  $\Delta_s$  decreases exponentially with increasing Z at  $Z > Z_s$ .

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Translated by J. G. Adashko

# Effect of Penning collisions between optically oriented Rb and He atoms on electron density in plasma

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The effect of mutual spin orientation of Rb atoms and metastable  $(2^{3}S_{1})$  He atoms on electron density in plasma, due to the dependence of the free-electron yield during Penning collisions between Rb atoms and metastable He atoms on their mutual spin orientation, has been observed experimentally and investigated.

PACS numbers: 52.20.Hv

The effect of optical orientation of atoms on electron density in plasma was described in<sup>[11]</sup>, where the discovery of the variation in the electrical conductivity of helium plasma during optical orientation of metastable helium atoms was reported and investigated. Once it was established that the total spin was conserved in ionizing (Penning) collisions between metastable helium atoms, <sup>[21]</sup> the change in the electron density under the influence of optical orientation was attributed to the dependence of the free-electron yield during Penning collisions between the metastable orthohelium atoms on their mutual spin orientation.<sup>(3-51</sup> Conservation of total spin should cause the mutual spin orientation to affect the free-electron yield not only in the case of collisions of metastable helium atoms with one another, but also in the case of collisions between metastable helium atoms and alkali metal atoms.<sup>(61)</sup> This phenomenon is of considerable interest for the investigation of the spin dependence of Penning collisions since, in contrast to the case of collisions between metastable helium atoms with one another, it offers the possibility of independent variation of the spin orientation of the two colliding atoms.

The aim of this work was to observe and investigate experimentally the dependence of the electron density in a plasma on the mutual spin orientation of rubidium atoms in the ground  $5^2S_{1/2}$  state and metastable  $2^2S_1$  helium atoms (the latter will be denoted by He\*). The existence of this dependence follows from a consideration of the Penning ionization process

$$He(2^{3}S_{4}) + Rb(5^{2}S_{2}) \rightarrow He(1^{4}S_{0}) + Rb^{+}(4^{4}S_{0}) + e^{-}.$$
 (1)

This reaction is forbidden when total spin is conserved and the spins of the original atoms are parallel, because the total spin of the original atoms is then 3/2and the total spin of the reaction products is only 1/2. If the mutual orientation of the metastable Hc\* atoms and Rb atoms is changed, the reaction (1) becomes allowed, and this leads to an increase in the yield of free electrons which can be detected from the change in the electron density in the plasma.

### **1. EXPERIMENTAL METHOD**

The change in the electron density in the plasma was determined (as in<sup>[1]</sup>) from the change in the electrical conductivity of the plasma, which led to a change in the high-frequency voltage across the electrodes used to produce the discharge in the working cell. The cylindric cell (6 cm long, 4 cm in diameter) was filled with rubidium vapor at room temperature and helium at 0.5 Torr. A high-frequency discharge was produced in the cell with the aid of two external open-ring electrodes. The electrodes were connected in parallel with the inductance of a tank circuit tuned to 20 MHz and coupled to the high-frequency oscillator feeding the discharge in the cell. The high-frequency voltage between the cell electrodes was measured with a diode amplitude detector. The detector time constant was 10  $\mu$ sec. The alternating component of the signal across the detector load was fed through a source follower to a narrow-band amplifier. A synchronous or amplitude detector was connected to the amplifier output. After detection, the signals were recorded with an x-yrecorder.

The optical orientation of the rubidium and helium atoms in the cell was achieved with the aid of two circularly polarized beams of light traveling in opposite directions and produced by rubidium and helium pump lamps. The light was polarized by polaroid films and mica quarter-wave plates. The sign of the circular polarization could be measured by rotating the mica plate through 90°. Helmholtz coils were used to produce the constant magnetic field  $H_0$ . This field was always parallel to the pump beam. Two independent pairs of coils with axes perpendicular to the field  $H_0$ were used to produce the radiofrequency magnetic fields.

All the experiments were performed with the weakest possible discharge in the cell, since the observed signals decreased rapidly with increasing strength of the discharge. The amplitude of the high-frequency voltage between the cell electrodes was usually 25 V.



FIG. 1. Signals representing the variation in the electrical conductivity of a plasma: (a) magnetic resonance in the ground  $5^{2}S_{1/2}$  state of Rb<sup>85</sup> atoms ( $f_0 = 290$  kHz); (b) magnetic resonance in the metastable  $2^{3}S_{1}$  state of He<sup>4</sup> ( $f_0 = 1760$  kHz).

The mutual spin orientation of the Rb and He\* atoms was varied by a radiofrequency resonance magnetic field, by changing the sign of the circular polarization of the pump beam, and by modulating the pump intensity at magnetic resonance.

### 2. EXPERIMENTAL RESULTS

Figure 1 shows signals illustrating the variation in the electrical conductivity of a plasma during magnetic resonance in the ground  $5^2S_{1/2}$  state of Rb<sup>85</sup> atoms (Fig. 1a) and the metastable  $2^{3}S_{1}$  state of helium atoms (Fig. 1b). The signals shown in Fig. 1 were obtained by differential passage through resonance, using low-frequency (333 Hz) modulation of the magnetic field  $H_0$ . These signals are therefore the derivatives of the absorption curves. In the case of signals marked 1 in Figs. 1a and b, the pump beams from the rubidium and helium lamps had circular polarizations of opposite direction. The spin moments of Rb and He\* were aligned in the same direction. Signal 2 in Fig. 1a and signal 3 in Fig. 1b were recorded with the direction of the circular polarization of the beam from the rubidium lamp reversed. The spin moments of Rb and He\* were then aligned in opposite directions. Signal 1 in Fig. 1a and all the signals in Fig. 1b correspond to a reduction in the high-frequency voltage between the cell electrodes and, consequently, to an increase in the electron density in the plasma at resonance. Signal 2 in Fig. 1a corresponds to a reduction in the electron density. Signal 2 in Fig. 1b was recorded with the rubidium lamp switched off. No signals were observed when the helium lamp was switched off and the rubidium lamp switched on.

It is thus clear from Fig. 1 that the electron density in the plasma does, in fact, depend on the mutual spin orientation of the Rb and He atoms. Figure 1 also shows that (1) the electron density undergoes a resonant change during passage through the magnetic resonance for both the Rb and He\* atoms, (2) when the circular polarization of the pump light from the rubidium lamp is reversed, the electrical-conductivity signal from the plasma changes sign at the magnetic resonance of the Rb atoms (signals 1 and 2 in Fig. 1a) and undergoes a change in size (without change of sign) at the magnetic resonance of the He\* atoms (signals 1 and 3 in Fig. 1b), and (3) when the He\* atoms pass through magnetic resonance, the electrical-conductivity signal from the plasma is present even when the rubidium lamp is switched off (signal 2 in Fig. 1b), but the size of this signal is substantially smaller than when both the rubidium and helium lamps are switched on (signals 1 and 3 in Fig. 1b).

The data in Fig. 1 can be explained as follows. When the spin moments of Rb and He\* have the same direction, the probability of Penning ionization is low and the disorientation of the Rb atoms at magnetic resonance leads to an increase in the ionization probability and in the number of electrons in the plasma (signal 1 in Fig. 1a). When the spin moments of Rb and He\* point in opposite directions, this corresponds to high ionization probability. The disorientation of the Rb atoms in this case leads to a reduction in the ionization probability and in the number of electrons in plasma (signal 2 in Fig. 1a). It follows that magnetic resonance of Rb atoms may lead to either an increase or a reduction in the electron density in plasma, depending on the relative direction of the two circular polarizations.

The behavior of electron density should be the same in the case of the magnetic resonance of the He\* atoms. However, it was found experimentally that, in this case, there is only an increase in the electrical conductivity of plasma (signals 1 and 3 in Fig. 1b). This is so because, in the case of the magnetic resonance of the He\* atoms, the observed signals contain contributions from not only the Penning collisions between Rb atoms and the He\* metastables, but also collisions of the helium metastables with one another. Signal 2 in Fig. 1b, which was obtained with the rubidium pump switched off, illustrates the relative contribution of such collisions. As in<sup>[1]</sup>, this signal is due exclusively to Penning collisions between orthohelium metastables. Figure 1b shows that the contribution of He\*-He\* collisions to the signals is much greater than that due to Rb-He\* collisions, and corresponds to an increase in the electron density at resonance. The contribution of He\*-He\* collisions to the signal is independent of the polarization of the rubidium pump radiation. It follows that when the direction of the circular polarization of the pump light is reversed, there is a change in only the size but not the polarity of the signals.

At the point of magnetic resonance in the experiments described above, the macroscopic spin moment of the atoms rotated with the Larmor frequency about the magnetic field  $H_0$  at a certain angle to it, and was reduced in magnitude (this is the radiofrequency saturation effect). Consequently, the plasma conductivity signals shown in Fig. 1 are connected both with the change in the angle between the macroscopic spins of Rb and He\*, and with the change in the size of one of the spins. In order to isolate the dependence of the electron density in plasma on the angle between the macroscopic spins of Rb and He\*, we carried out an experiment in which the pump light was modulated at



FIG. 2. In-phase (1) and quadrature (2) components of the signal representing the variation in the electrical conductivity of plasma with the rubidium pump modulated under the conditions of magnetic resonance in the ground  $5^2S_{1/2}$  state of Rb<sup>85</sup> atoms ( $f_m = 4$  kHz,  $f_0 = 290$  kHz,  $H_0 = 0.621$  Oe).

magnetic resonance. It is well known<sup>[7]</sup> that modulation of the pump light and the point of magnetic resonance leads to an undamped rotation (with a nutation frequency  $\omega_1 = \gamma H_1$ ) of the macroscopic spin moment about the direction of the radiofrequency resonance magnetic field  $H_1$ , provided the light modulation frequency is equal to the nutation frequency.

Figure 2 shows signals representing the variation in the electrical condictivity of the plasma when the intensity of the rubidium pump was modulated in the case of magnetic resonance in the ground  $5^2S_{1/2}$  state of Rb<sup>85</sup> atoms. The modulation frequency  $\omega_m/2\pi$  was 4 kHz. The signals in Fig. 2 were recorded using narrow-band amplification at 4 kHz and synchronous detection. The scan was obtained by slow variation of the amplitude of the resonance radiofrequency magnetic field  $H_1$ . Signals 1 and 2 in Fig. 2 were obtained at different phases of the reference voltage in the synchronous detector (these phases differed by 90°).

Figure 2 shows that the amplitude of the electrondensity oscillations undergoes a resonance change near  $H_1 = H_{10}$ . The measured value of  $H_{10}$  was 8.5 mOe. The nutation frequency  $\omega_1/2 = (\gamma/2\pi)H_1$  (for Rb<sup>85</sup>, we have  $\gamma/2\pi = 0.47$  kHz/Oe) for this value of  $H_1$  is 4 kHz, i.e., it is equal to the modulation frequency of the Rb pump.

In the experiment using the modulation of the pump light at magnetic resonance, effects connected with the change in the mutual orientation are separated from those associated with the magnitude of the macroscopic spins. The electron-density oscillation with frequency  $\omega_m$  at fixed  $H_1$  is connected only with the change in the angle between the macroscopic spin moment of the Rb atoms and the spin of the He\* atoms (this angle varies at the rate  $\omega_m$ ). The variation in the amplitude of oscillations of the electron density with varying  $H_1$ , on the other hand, is connected only with the change in the size of the macroscopic moment of the Rb atoms.

In the experiments described above, we varied only one of the macroscopic spins (of the Rb or He\* atoms). In another experiment, described below, both spins (Rb and He\*) were varied both in magnitude and direction. In this experiment, we achieved simultaneous magnetic resonance of the He\* and Rb atoms. Two independent pairs of coils were used to produce in the cell two resonance radiofrequency magnetic fields with frequencies  $f_0(\text{He}^*)$  and  $f_0(\text{Rb}^{85})$ . These frequencies were equal to the Larmor frequencies of He\* and  $\text{Rb}^{85}$  in a field  $H_0 = 17$  mOe. The amplifier was tuned to the difference frequency  $\Delta f = f_0(\text{He}^*) - f_0(\text{Rb}^{85}) = 40$  kHz. An



FIG. 3. Signal showing the variation in the electrical conductivity of plasma for the difference frequency  $\Delta f = f_0$  (He<sup>\*</sup>)  $-f_0$  (Rb<sup>85</sup>) in the case of simultaneous magnetic resonance of the Rb<sup>85</sup> and He<sup>\*</sup> atoms [ $f_0$  (He<sup>\*</sup>) = 48.4 kHz,  $f_0$  (Rb<sup>85</sup>) = 8.1 kHz].

amplitude detector was connected to the amplifier output.

Figure 3 shows the plasma electrical-conductivity variation signal obtained in this experiment. The relative change in the high-frequency voltage between the cell electrodes, which corresponds to the signal in Fig. 3, was  $2 \times 10^{-7}$ . For comparison, we note that, for the signals shown in Fig. 1a, this change was 2  $\times 10^{-6}$ . During the simultaneous magnetic resonance of the Rb and He\* atoms, the macroscopic spin moments of these atoms rotate about the direction of the field  $H_0$ with their own Larmor frequencies. The magnitude of these moments (for fixed  $H_0$ ) does not change, but the angle between them varies with at the rate  $\Delta \omega = 2\pi \Delta f$  $=2\pi [f_0(\text{He}^*) - f_0(\text{Rb}^{85})]$ . The data shown in Fig. 3 indicate that the electron density oscillates with the frequency  $\Delta f = \Delta \omega / 2\pi$ , and the amplitude of these oscillations changes in a resonant fashion near the point  $H_0$  $=H_{p}$ , which corresponds to the simultaneous resonance of Rb and He\*. As in the previous case, this experiment shows the separation of effects connected with the variation in the angle between the spins and with the variation in the magnitude of the spins.

It is important to note that the signals representing the variation in the electrical conductivity of plasma in the case of the difference frequency  $\Delta f$  could be observed only in very weak magnetic fields (in the case of Fig. 3,  $H_p = 17$  mOe). An increase in the magnetic field (and, correspondingly, an increase in the Larmor frequency of Rb and He\*) resulted in a reduction in the size of these signals. They vanished altogether for  $H_0$ = 100 mOe ( $\Delta f = 250$  kHz).

#### 3. DISCUSSION

The results of the foregoing experiments show that the mutual spin orientation of the rubidium and metastable orthohelium atoms has an effect on the electron density in plasma. The reason for this is that the freeelectron yield of Penning collisions between Rb and He\* atoms depends on their mutual spin orientation [reaction (1)], i.e., that the cross section  $\sigma$  for Penning ionization depends on the spin orientation of the colliding atoms. Since the cross section is an average over all the possible collisions between Rb and He\* atoms, it should be expressed in terms of the macroscopic (averaged over the ensemble of atoms) spin moments of the Rb and He\* atoms:

$$\sigma = F(\langle \hat{\mathbf{S}}_{\mathrm{He}} \rangle, \langle \hat{\mathbf{S}}_{\mathrm{Rb}} \rangle).$$
(2)

The kinetic equation for the electron density  $n_s$  in the plasma can be written in the form

$$dn_c/dt = -n_e/\tau + v\sigma N_{\rm He} \cdot N_{\rm Rb} + I, \qquad (3)$$

where  $\tau$  is the average effective lifetime of the electrons in the plasma, v is the average relative velocity of the Rb and He\* atoms,  $N_{\rm Rb}$  and  $N_{\rm He*}$  are the densities of these atoms,  $(v\sigma N_{\rm He*}N_{\rm Rb})$  is the rate at which the electrons are produced as a result of Rb-He\* collisions, and I is the rate at which they are produced in other processes.

Equation (3) for the electron density  $n_e$  is linear and, therefore, the electron density should be a linear function of  $\sigma$ . When the variation in the electron density is small, the change in the voltage across the cell electrodes should be directly proportional to the change in the electron density and, consequently, directly proportional to the change in the cross section for Penning ionization. The small size of the signals observed in the experiment suggests that the changes in the electron density were also small, and that the observed signals were directly proportional to the change in the cross section  $\sigma$ . Consequently, the spin dependence of the cross section for Penning ionization, i.e., the explicit form of the function (2), can, at least in principle, be established from the observed dependence of the signals upon the macroscopic values of the spin moments  $\langle \hat{\mathbf{S}}_{Rb} \rangle$ and  $\langle \hat{\mathbf{S}}_{\mathrm{He}} \rangle$  of the Rb and He\* atoms.

Let us now list the main observed properties of this dependence:

(1) the signals change their polarity when the direction of  $\langle \hat{\mathbf{S}}_{Rb} \rangle$  or  $\langle \hat{\mathbf{S}}_{H\bullet^*} \rangle$  is reversed. (2) the signals depend on  $|\langle \hat{\mathbf{S}}_{Rb} \rangle|$  and  $|\langle \hat{\mathbf{S}}_{H\bullet^*} \rangle|$ , and (3) variation of the angle between  $\langle \hat{\mathbf{S}}_{Rb} \rangle$  and  $\langle \hat{\mathbf{S}}_{H\bullet^*} \rangle$  at the rate  $\omega$  leads to the appearance of signals of frequency  $\omega$ . Since in (2) the scalar  $\sigma$  is a function of the two vectors  $\langle \hat{\mathbf{S}}_{H\bullet^*} \rangle$  and  $\langle \hat{\mathbf{S}}_{Rb} \rangle$ , it follows that, in view of the measured properties of this function, the cross section  $\sigma$  is a linear function of the scalar product of these two vectors:

$$\sigma = A + B \langle \hat{\mathbf{S}}_{\text{He}} \rangle \langle \hat{\mathbf{S}}_{\text{Rb}} \rangle.$$
(4)

where A and B are independent of the spin moments.

The above experimental data do not, however, enable us to carry out a quantitative verification of expression (4). On the other hand, this expression agrees with all the properties of the experimentally obtained signals listed above.

Let us now consider the effect of the rate of change of the cross section  $\sigma$  on the variation of the electron density. Let us suppose that  $\sigma = \sigma_0 + \sigma_m \cos \omega t$ . The solution of (3) for  $t \gg \tau$  then takes the form

$$n_{e} = (I + \upsilon \sigma_{0} N_{He} \cdot N_{Rb}) \tau + \upsilon \sigma_{m} N_{He} \cdot N_{Rb} \frac{\tau}{\left[1 + (\omega \tau)^{2}\right]^{\gamma_{h}}} \cos\left(\omega t - \varphi\right), \qquad (5)$$

where  $\varphi = \arctan(\omega \tau)$ . It follows from this formula that, when  $\omega \tau \gg 1$ , the amplitude of the variation in the electron density is inversely proportional to the frequency  $\omega$  of the cross section  $\sigma$ . The electron lifetime  $\tau$  at low pressures is determined by the time of diffusion to the cell walls. The value of  $\tau$  estimated as in<sup>[8]</sup>, turns out to be  $\tau = 1.5$  $\times 10^{-5}$  sec under our conditions. This means that  $\omega \tau = 1$ corresponds to  $\omega/2\pi = 11$  kHz. Consequently, when the mutual orientation (or magnitude) of the spin moments of Rb and He\* varies with a frequency exceeding 11 kHz, the signals corresponding to the variation in the electron density should decrease with increasing frequency. This appears to explain the fact that signals such as those shown in Fig. 3 were not observed in strong magnetic fields.

We note in conclusion that, even a partial conservation of the resultant spin during Penning collisions between Rb and He\* suffices to explain the experimental dependence of the mutual spin orientation of the Rb and He\* atoms on the electron density in the plasma. To assess the extent to which the resultant spin is conserved during such collisions requires further investigation.

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Translated by S. Chomet

# Gas diffusion: Its dependence on nuclear spin

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The influence of the nuclear spin on the diffusion of a molecular gas due to the precession of the angular momentum in the electric field of the nuclear quadrupole or in the magnetic field of the nuclear magnetic moment is considered. The expected effect could be detected at low pressure in gas mixtures with molecules of the type HA, where among the isomers of the nucleus A there are long lived isomers with large nuclear spins. In principle this effect could be used for the separation of nuclear isomers.

PACS numbers: 51.10.+y

1. The theory of gas diffusion allows one to predict the change in the diffusion coefficient of a molecular gas in the case when the nuclear spin of one of the atoms which make up the molecule differs from zero, as compared with the diffusion coefficient of isomers of the same molecules without nuclear spin. This effect is analogous to the well-known Senftleben effect (cf. the reviews<sup>[1]</sup>) consisting in the influence of an external magnetic field on the kinetic coefficients of molecular gases.

The gist of the effect is related to the fact that the nuclear spin violates the conservation of the angular momentum **K** of the molecule during its free flight between collisions. In the absence of nuclear spin the diffusion coefficient is proportional to the mean free path  $\lambda$ , averaged over the different orientations of the angular momentum of the molecule. In the presence of spin the molecule tumbles and its angular momentum is not constant along the free path. Therefore one first averages the scattering cross section  $\sigma$  over the directions of the angular momentum and then calculates the mean free path from this average cross section. If the cross section depends on the orientation of the angular momentum then the difference between  $\overline{\lambda} \propto \overline{\sigma^{-1}}$  and  $\lambda$ 

 $\propto (\overline{\sigma})^{-1}$  may be substantial. We consider this result of influence of the nuclear spin on diffusion using the diffusion of diatomic molecules in a monoatomic gas as an example.

2. The diffusion of molecules in an atomic gas is described by a kinetic equation which is classical in the translational degrees of freedom and quantized in the internal degrees of freedom:

$$\partial f/\partial t + (\mathbf{v}\nabla)f + i\hbar^{-1}[H, f] = -J_{\text{coll}}$$
 (1)

Here f is the molecular distribution function and  $J_{coll}$  is the collision integral. The operator describing the internal energy of the molecule will be written in the form<sup>[2]</sup>

$$H = B\hat{\mathbf{K}}^2 + V. \tag{2}$$

where the first term describes the rotational energy of the molecule and the second term determines the hyperfine interaction between the rotation of the molecules and the nuclear spin. For definiteness we consider a nonparamagnetic diatomic molecule in which only one of the nuclei has nonzero spin *I*. If  $I \ge 1$  the main contribution to *V* comes from the quadrupole interaction of the nucleus with the electrons. If  $I = \frac{1}{2}$  the source