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ABSTRACT

The radiofrequency paramagnetic-resonance method was used to observe the exchange collisions between the singly ionized Xe ground state \(^2P_{3/2}\) and the neutral metastable state \(^3P_2\). The ionic ground-state and metastable-state atoms are both formed and aligned by unidirectional low-energy high-flux electron-beam impact. The magnetic resonances of even and odd ionic ground-state atoms were observed by monitoring changes in the transparency of the resonance radiation absorbed by the metastable-state atoms.

Phenomenological theories are presented which explain the sign of the magnetic resonance signal of the ionic ground state relative to that of the neutral metastable state, together with independent experimental evidence to support the theories. Conclusive experimental evidences is presented supporting our original assumptions that the ionic ground state Xe\(^+\)\(^(2P_{3/2})\) is aligned by low-energy electron-impact ionization and that such alignment is detectable through results of the exchange collisions with the metastable state.
I. INTRODUCTION

Atoms excited by unidirectional (i.e., parallel) electron beams are, in general, aligned. For aligned excited states that decay by spontaneous emission of light, resonance radiations are therefore partially polarized. Magnetic resonances of excited states that cause alignment reduction also reduce the polarization ratio of the emitted light. On this basis, various properties of the excited states such as fine-structure separations, hyperfine-structure separations, $g_J$, $g_F$, and lifetimes have been measured.

For metastable states formed and aligned by electron-impact excitation, the same atomic properties as for the radiative excited states can be measured by monitoring the change in transparency of the resonance radiations absorbed by the metastable-state atoms. Recently we reported that the ionic ground state of Xe$^+$(2P$_{3/2}$) atoms, which is difficult to observe, can be studied optically by this metastable-state resonance-absorption method through collision-induced coupling with metastable-state atoms. In this paper, we present more detailed information describing the observed phenomena.

A high-flux low-energy unidirectional electron beam was produced by a space-charge-neutralization technique using low-pressure Xe gas as a source of ions to neutralize the electron space charge. By this method, aligned ionic ground-state atoms at relatively high density, as well as metastable-state atoms, are formed. In a steady-state condition, a certain population inequality results for Xe$^+$(2P$_{3/2}$) and Xe(3P$_2$) magnetic sublevels due to alignment by electron impact and through subsequent spin-orbit coupling, ionic ground-state–metastable-state collisions,
collisions with the ground state of odd Xe isotopes, and the spin-lattice relaxation due to collision with walls, foreign gas atoms, and plasma discharge products. However, we consider only the contribution to steady-state conditions that is due to the ionic ground-state-metastable-state collisions, since we can set our equipment to be sensitive to only this process, all other processes appearing as shortening of the spin-lattice relaxation time.

The magnetic resonance in the ionic ground state that equalizes the population distribution redistributes the population of the metastable state through subsequent exchange collisions. Thus the changes in transparency of the resonance radiations that are absorbed by the metastable state allow us to observe rf resonance of the ionic ground state. The ionic-ground-state resonance signal is compared with the magnetic resonance signal of the metastable state, and one finds the relative signs of the signals are opposite.

II. PHENOMENOLOGICAL THEORY

The relevant energy-level diagram is shown in Fig. 1. By electron impact the magnetic sublevels \( M_J = 0 \) and \( \pm 1 \) of Xe\((^3P_2)\) are more excited than \( M_J = \pm 2 \) states at slightly above the threshold energy of \(^3P_2\). We say that such an excited state is aligned. Although no theory is available at present dealing with the selective excitation of Zeeman magnetic sublevels by electron-impact ionization, we expect that the ionic ground state \( \text{Xe}^+\left(^2P_{3/2}\right) \) is aligned. However, since we expect neither threshold-energy conditions nor available theories to be rigorously satisfied, we assign the production rates \( \alpha, \beta, \) and \( \gamma \) for the...
magnetic sublevels \( M_J = \pm 2, \pm 1, \) and 0 of \( \text{Xe}(^3P_2) \). Similarly, we assign the production rates \( \alpha' \) and \( \beta' \) for the magnetic sublevels \( M_J = \pm 3/2 \) and \( \pm 1/2 \) of \( \text{Xe}^+(^2P_{3/2}) \). Since the electron impact aligns only the excited atoms, the production rate is the same for \( M_J \) and for \(-M_J\).

The metastable-state and ionic ground-state spin-relaxation times \( T_R \) and \( T_I \) represent the process that tends to cause the magnetic sublevel population to reach its thermal-equilibrium distribution by collisions with the container wall, electrodes, and impurities, etc., excluding the process involving interaction with metastable ionic ground-state collisions.

Coupling between the metastable-state atoms and the ionic ground-state atom is induced through collisions. Equilibrium between the two systems is established in the characteristic time \( \tau \approx 1/\sigma v n \) for the metastable state and \( \tau' \approx 1/\sigma v N \) for the ionic ground state, where \( \sigma, v, N, \) and \( n \) are respectively the total exchange cross section (spin exchange and spin exchange associated with charge exchange), relative velocity, metastable-state atom density, and ionic ground-state atom density.

In addition to ionic ground-state–metastable-state collisions, we should consider the exchange collisions between metastable state and \(^4S_0\) isotopic atoms whose nuclear spins are not zero, as well as collisions of atoms having the same configuration. Such a phenomenon results in the alignment of nuclei having nuclear spin \( I > \frac{1}{2} \) in the \(^4S_0\) ground state.\(^6\) For simplicity, we include such processes in \( T_R \) and \( T_I \). We further assume that, due to collisions between \( \text{Xe}^+(^2P_{3/2}) \), the angular
momentum changes at most one unit per collision. This assumption is a reasonable one, since we believe the dominant mechanism is due to electron spin exchange.

Let \( N_2, N_1, N_0, N_{-1}, \) and \( N_{-2} \) be the population densities of the magnetic sublevels of \( \text{Xe}(3\,P_2) \) corresponding to \( M_J = 2, 1, 0, -1, \) and \( -2. \) Similarly, let \( n_{3/2}, n_{1/2}, n_{-1/2}, \) and \( n_{-3/2} \) be the population densities of the magnetic sublevels of \( \text{Xe}^+(2\,P_{3/2}) \) corresponding to \( M_J = 3/2, 1/2, -1/2, \) and \( -3/2. \) From these assumptions, we construct the following rate equations:

\[
\frac{dN_2}{dt} = - \sum_{j \neq 2} N_j W_{j,2} + \sum_{j \neq 2} N_j W_{j,2} - \frac{1}{\tau} N_2 + \alpha + v\sigma [N_1 \frac{n}{2} - N_2 (n-n_{3/2})],
\]

\[
\frac{dN_1}{dt} = - \sum_{j \neq 1} N_j W_{j,1} + \sum_{j \neq 1} N_j W_{j,1} - \frac{1}{\tau} N_1 + \beta + v\sigma [-N_1 n + N_2 (n-n_{3/2})]
+ N_0 \frac{n}{2},
\]

\[
\frac{dN_0}{dt} = - \sum_{j \neq 0} N_j W_{j,0} + \sum_{j \neq 0} N_j W_{j,0} - \frac{1}{\tau} N_0 + \gamma + v\sigma [N_{1/2} + N_{-1/2}]
- N_0 n],
\]

\[
\frac{dN_{-1}}{dt} = - \sum_{j \neq -1} N_j W_{j,-1} + \sum_{j \neq -1} N_j W_{j,-1} - \frac{1}{\tau} N_{-1} + \beta + v\sigma [-N_{-1} n + N_{-2} (n-n_{3/2})]
+ N_0 \frac{n}{2}].
\]
\[
\frac{\text{d}N_{-2}}{\text{d}t} = - \sum_{j' \neq -2} N_{-2} W_{-2, j} + \sum_{j' \neq -2} N_{j} W_{j, -2} - \frac{4}{\tau} N_{-2} N_{1} n_{2}^{\frac{1}{2}} N_{-2} \left( n_{-3/2} - n_{3/2} \right),
\]

\[
\frac{\text{d}n_{3/2}}{\text{d}t} = - \sum_{j' \neq 3/2} n_{3/2} w_{3/2, j} + \sum_{j' \neq 3/2} n_{j} w_{j, 3/2} - \frac{4}{\tau'} n_{3/2} + \alpha' + \nu \sigma\left[ -n_{3/2} (N - N_{2}) + n_{1/2} \frac{N}{2} \right],
\]

\[
\frac{\text{d}n_{1/2}}{\text{d}t} = - \sum_{j' \neq 1/2} n_{1/2} w_{1/2, j} + \sum_{j' \neq 1/2} n_{j} w_{j, 1/2} - \frac{4}{\tau'} n_{1/2} + \beta' + \nu \sigma\left[ n_{3/2} (N - N_{2}) - n_{1/2} N + n_{-1/2} \frac{N}{2} \right],
\]

\[
\frac{\text{d}n_{-1/2}}{\text{d}t} = - \sum_{j' \neq -1/2} n_{-1/2} w_{-1/2, j} + \sum_{j' \neq -1/2} n_{j} w_{j, -1/2} - \frac{4}{\tau'} n_{-1/2} + \beta' + \nu \sigma\left[ n_{1/2} \frac{N}{2} - n_{-1/2} N + n_{-3/2} (N - N_{2}) \right],
\]

\[
\frac{\text{d}n_{-3/2}}{\text{d}t} = - \sum_{j' \neq -3/2} n_{-3/2} w_{-3/2, j} + \sum_{j' \neq -3/2} n_{j} w_{j, -3/2} - \frac{4}{\tau'} n_{-3/2} + \alpha' + \nu \sigma\left[ n_{-1/2} \frac{N}{2} - n_{-3/2} (N - N_{2}) \right],
\]

where \( N = \sum_{j' = -2}^{2} N_{j} \) and \( n = \sum_{j' = -3/2}^{3/2} n_{j} \).

\( \tau \) and \( \tau' \) are respectively the life-time of the \( \text{Xe}(^3P_2) \) and \( \text{Xe}^+(^2P_{3/2}) \).
state, and $W_{ij}$ and $w_{kl}$ are spin relaxation rate of the metastable and ionic ground-state respectively.

The first-order steady-state solutions are given as

$$N_2^{(1)} = N_{-2}^{(1)} = \frac{1}{5 + \frac{1}{\tau_w}} \left\{ N + \frac{\alpha}{W} + \frac{\gamma\sigma}{W} \left[ \frac{1}{2} N_2^{(0)}(n - n_3^{(0)}) + \frac{n}{2} N_1^{(0)} \right] \right\} ,$$

$$N_1^{(1)} = N_{-1}^{(1)} = \frac{1}{5 + \frac{1}{\tau_w}} \left\{ N + \frac{\beta}{W} + \frac{\gamma\sigma}{W} \left[ N_2^{(0)}(n - n_3^{(0)}) - N_1^{(0)} n + \frac{1}{2} N_0^{(0)} n \right] \right\} ,$$

$$N_0^{(1)} = \frac{1}{5 + \frac{1}{\tau_w}} \left\{ N + \frac{\gamma}{W} + \frac{\gamma\sigma}{W} \left[ N_1^{(0)} n - N_0^{(0)} n \right] \right\} ,$$

where $N_k^{(0)}$ and $n_{kl}^{(0)}$ are the zero-order solutions given by

$$N_2^{(0)} = N_{-2}^{(0)} = \frac{N + \alpha/W}{5 + \frac{1}{\tau_w}} ,$$

$$N_1^{(0)} = N_{-1}^{(0)} = \frac{N + \beta/W}{5 + \frac{1}{\tau_w}} ,$$

$$N_0^{(0)} = \frac{N + \gamma/W}{5 + \frac{1}{\tau_w}} ,$$

$$n_{3/2}^{(0)} = n_{-3/2}^{(0)} = \frac{n + \alpha'/w}{4 + \frac{1}{\tau_w}} ,$$

$$n_{1/2}^{(0)} = n_{-1/2}^{(0)} = \frac{n + \beta'/w}{4 + \frac{1}{\tau_w}} .$$

Here, for simplicity, we assumed $W = W_{ij}$ and $w = w_{kl}$; i.e., we assumed a uniform relaxation rate.

For simplicity, we consider the case in which the electron beam direction is parallel to the externally applied static field $H_0$. Then the maximum obtainable magnetic resonance signals of the $\text{Xe}^+ (^2 \text{P}_{3/2})$ ionic...
ground state observed through resonance absorption by the $\text{Xe}(^3\text{P}_2)$
neutral metastable state are given for transition $J = 2$ and $J = 1$
(see Table I) by

$$\text{Signal}_\parallel \propto 8 (\Delta N_0) + 2 (6) (\Delta N_1)$$

$$\approx -12 \frac{N + \alpha/W}{(5 + 1/W)} (\frac{n}{4} - n_{3/2}^{(0)}) \cdot \frac{\nu \sigma}{W} \approx -\frac{3}{50} \frac{\beta' - \alpha'}{wW} N \nu \sigma,$$  \hspace{1cm} (19)

$$\text{Signal}_\perp \propto 2 (1) (\Delta N_0) + 2 (3) (\Delta N_1) + 2 (6) (\Delta N_2) \approx \frac{3}{100} \frac{\beta' - \alpha'}{W} N \nu \sigma,$$  \hspace{1cm} (20)

where $\Delta N_j = (N_j n_{3/2} - (N_j)_{\text{steady state}}$.

The metastable-state resonance signals are given to zero order
by

$$\text{Signal}_\parallel \propto -\frac{4}{5W} (\gamma + \beta - 2\alpha)$$  \hspace{1cm} (21)

and

$$\text{Signal}_\perp \propto \frac{2}{5W} (\beta + \gamma - 2\alpha)$$  \hspace{1cm} (22)

Our previous experiment showed that $\alpha < \gamma, \beta$; i.e., the states
$M_J = 0$ and $\pm 1$ of $\text{Xe}(^3\text{P}_2)$ are selectively more excited than those with
$M_J = \pm 2$. From Eq. (21), we find that the resonance absorption decreases
due to the metastable-state paramagnetic resonance. Now, from Eq. (19)
we find that the relative signs of the signals between $\text{Xe}^+ (^2\text{P}_{3/2})$ and
$\text{Xe}(^3\text{P}_2)$ magnetic resonance depend on $\beta' - \alpha'$. If $\beta' - \alpha' > 0$, the
relative signs of the signals are the same; and if $\beta' - \alpha' < 0$, the relative
signs of the signals are unlike, resulting in an increase in resonance ab-
sorption by the metastable state due to the $\text{Xe}^+ (^2\text{P}_{3/2})$ ionic ground-state
magnetic resonance. We experimentally observed that the latter is the case,
indicating the very interesting phenomenon that the $M_J = \pm 3/2$ states of $\text{Xe}^+(2^{3/2}P_{3/2})$ are more excited by low-energy electron impact ionization than the $M_J = \pm 1/2$. Figure 2 shows the diagrammatic representation of the phenomenon. This phenomenon, at first glance, may seem somewhat surprising in view of Dehmelt-type spin-exchange phenomena, in which a mixture of two species of atoms is placed in a cell and one of the species is oriented by optical pumping. The collisions result in the orientation of the second species, so that the orientations of both species have the same sign. Destruction of the orientation of either one of the species results in destruction of the orientation of the other.

Now, let us consider what would happen if one could orient these two species of atoms opposite each other by some means, for example, by two pumping lights. If the pumping speeds were higher than the spin relaxation rates due to spin-exchange collisions, in the steady states, both species would still be partially oriented, opposite to each other. Now, if one caused the magnetic resonance of one of the species, it is obvious that the degree of orientation of the other species would increase. We shall show the experimental evidence of this phenomenon in the latter part of this paper. Now, our experimental situation is quite analogous to what we have just described, except that we deal with an alignment instead of orientation. However, this alters the discussion only slightly.
III. EXPERIMENTAL CONSIDERATIONS

Production of a High-Flux Electron Beam

In order to obtain a detectable signal, the following conditions should be met:

(a) The electron beam should be parallel; therefore, gas pressure should be low to avoid multiple scattering of the electron beam.
(b) The electron-beam current density should be high.

The maximum electron current that can flow from a "hot" cathode to an anode in high vacuum is limited by the space charge of the electron, so that the current under such conditions is too small to produce a sufficient concentration of atoms in the metastable and ionic ground states. However, from Langmuir's theory of space-charge-neutralized electron flow, we know that if even a small amount of gas is present and the applied voltage is higher than the ionization potential, the positive ions formed tend to neutralize the electron space charge, and thus allow the current to increase until it is limited only by the electron emission of the cathode, which depends upon the cathode temperature. Under conditions of space-charge neutralization, the cathode is surrounded by the ion sheath, and the rest of the volume is filled by nearly field-free plasma. The voltage difference between the cathode and anode is concentrated between the cathode and ion sheath, so that the electrons receive most of their acceleration between the cathode and sheath, and enter the relatively field-free plasma perpendicular to the cathode surface.

Thus, by the space-charge-neutralization method, the requirement of unidirectional high-electron current can be satisfied.
very ions that space-charge neutralize are formed by unidirectional electron impact, we expect selective excitation of Zeeman magnetic sublevels, just as the magnetic sublevels of the neutral metastable-state atoms are selectively excited.

The experimental arrangements have been described in our previous papers, with modified signal detection methods for detecting the extremely small signals arising from odd isotopes of Xe\(^+\)(^2P_{3/2})\).\(^{3,4,6}\) However, we wish to mention that it is not necessary to use an external light source, since the light radiated from the plasma in the absorption cell itself acts as light source, as is described in our previous paper.

IV. EXPERIMENTAL RESULTS AND ANALYSIS

Figures 3 and 4 show the effects of optical absorption in the cell as a function of the applied magnetic field. The signals in the peak correspond to the Xe\(^+(^3P_2)\) metastable state and the signals in the trough correspond to \(g_J = 1.334\); we assign them to be due to the even isotope of the Xe\(^+\)(^2P_{3/2}) ionic ground state.

Because of the complexity of the system involved in this experiment (in contrast with more conventional types of experiments in which most of the atoms are in the ground state), we must be careful not to make an accidental misidentification of \(g_J = 1.334\) resonances that may originate from other excited states. We use the following arguments to assign the \(g_J = 1.334\) magnetic resonance to the Xe\(^+\)(^2P_{3/2}) ionic ground state:

(a) The Xe\(^+\)(^2P_{3/2}) ionic ground state has one electron missing from the closed shell. Therefore, we expect extremely good Russell-Saunders coupling, so that the \(g_J\) of Xe\(^+\)(^2P_{3/2}) should be very close to 4/3.

(b) The observed magnetic resonance band width is narrow, indicating
the relatively long spin-relaxation time, of the order of $10^{-5}$ sec. (Note that this is the spin-relaxation time, not the lifetime.) Therefore, the resonance must be due to the long-lived state.

(c) The magnetic resonance signals arising from the cascading transitions to the $^3P_2$ metastable state from the long-lived excited states that are initially aligned by direct electron impacts were experimentally observed and have been eliminated from the $g_J = 1.334$ resonance. Furthermore, a simple calculation, based on the $\Delta M_J = 0$ selection rule, shows that these magnetic resonances must have same sign as the magnetic resonances of the $^3P_2$ even and odd isotopes. Experimental observation showed that this is indeed the case. Since this observed phenomenon is not directly related to the main discussion in this paper, it will be reported in a separate paper.

(d) Finally, the most conclusive test is the observation of weak signals of odd isotopes of $^{129}$Xe$^+$(2$P_{3/2}$) and $^{131}$Xe$^+$(2$P_{3/2}$) (see Fig. 5). We observed that the signs of these signals are the same as the $g_J = 1.334$ resonance, and hence opposite to the metastable-state magnetic resonance signals. The accidental coincidence of $g_J = 1.334$ signals and $g_F$ signals associated with $g_J = 1.334$ with the same sign is extremely small, since $J$ values of neutral Xe are all integers, while those of the Xe II are all odd multiples of 1/2.

Next we consider the relative signs of the signals. Comparing the $^3P_2$ metastable-state signal of the even Xe isotope with Eq. (21), we observed experimentally that the magnetic sublevels of the $^3P_2$ metastable state with $M_J = 0$ and $\pm 1$ are more populated than those with $M_J = \pm 2$. With this information, comparison between Eqs. (21)
REFERENCES

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Table I. Relative absorption probabilities in transitions between magnetic sublevels $M_J$ of $^3P_2$ and $M'_J$ of $^3S_1$ of Xe.

<table>
<thead>
<tr>
<th>$M'_J$</th>
<th>$M_J$</th>
<th>Polarized light parallel to electron-beam direction</th>
<th>Polarized light perpendicular to electron-beam direction</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2</td>
<td>0</td>
<td>1/10</td>
</tr>
<tr>
<td>0</td>
<td>1</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>-1</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>-2</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
FIGURE LEGENDS

Fig. 1. Diagram of relevant energy levels of a neutral Xe atom and of a singly ionized Xe used in the experiment.

Fig. 2. (a) Steady-state population distribution before \( \text{Xe}^+ \left( ^2P_{3/2} \right) \) rf resonance.
(b) Effect of magnetic sublevel population redistribution of the \( \text{Xe} \left( ^3P_2 \right) \) metastable state due to \( \text{Xe}^+ \left( ^2P_{3/2} \right) \) magnetic resonance. The weights of the lines indicate the relative populations.

Fig. 3. Magnetic resonance of \( \text{Xe} \left( ^3P_2 \right) \) and \( \text{Xe}^+ \left( ^2P_{3/2} \right) \) with an rf frequency of 5.085 MHz.

Fig. 4. Magnetic resonance of \( \text{Xe} \left( ^3P_2 \right) \) and \( \text{Xe}^+ \left( ^2P_{3/2} \right) \) with an rf frequency of 3.400 MHz.

Fig. 5. Magnetic resonance of even and odd isotopes of \( \text{Xe}^+ \left( ^2P_{3/2} \right) \) ionic ground state. Modified fast data-accumulation technique employing digital to analog conversion logic for the field sweep was used.

Fig. 6. (a) Results of Dehmelt-type experiment in which only Cs is oriented by optical pumping.
(b) Result of magnetic resonance on Rb when the Cs and Rb are oppositely oriented: the Cs orientation is increased.
$Xe \rightarrow Xe^+$

$\lambda 8409$ linearly polarized light

$3S_1 \rightarrow 3P_2$

$2P_{3/2}$

"Spin-exchange" collisions

$(8.27 \text{ eV})$

$(12.13 \text{ eV})$

Electron-impact excitation

Fig. 1
Fig. 2

(a) Xe\(^+\) (\(^2P_{3/2}\))

(b) Xe\(^+\) (\(^2P_{3/2}\))

Collisions

Population density compression
Fig. 3
Magnetic resonance of metastable state ($^3P_2$) of Xe I even isotopes.

Magnetic resonance of ground state ($^2P_{3/2}$) of Xe II even isotopes.

Fig. 4
 rf resonance of $^{2}P_{3/2}$ of Xe$^+$ with $\iota = 3/2, F = 3, 2, 1$.
$\nu = 2.7927$ MHz.

- rf resonance of $^{2}P_{3/2}$ of $^{129}$Xe$^+$ with $\iota = 1/2, F = 1$.
$\nu = 6.1781$ MHz.

**Fig. 5**
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