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OBSERVATION OF SPIN EXCHANGE BETWEEN THE SINGLY IONIZED Xe^+ GROUND STATE AND THE METASTABLE STATE OF NEUTRAL XENON*

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We have observed spin-exchange collisions between the singly ionized $^2P_{3/2}$ ground state of Xe and the $^3P_2$ metastable state of neutral Xe, both formed and aligned by electronic-impact excitation under space-charge neutralization. Extension to the rf spectroscopy of the ionized ground state of other noble-gas atoms seems promising.

In this Letter we report what we believe is the first observation of spin-exchange collisions between the singly ionized ground state of Xe^+ and the metastable state of neutral Xe, both having P configurations. So far as we know, spin-exchange collision effects have been observed only between an oriented atom and an unoriented atom or an electron, all having zero orbital angular momentum ($L=0$). Dehmelt,[1] using a very elegant ion-storage collision technique, first succeeded in observing spin-exchange collisions between an optically pumped, oriented Cs atom and ionized $(He^4)^+$. This method was used quite recently on $(He^3)^+$, revealing an ultrahigh precession determination of hyperfine structure by this method.[2]

In our experiment, instead of an oriented atom colliding with an unoriented ion, both colliding atoms (Xe^+ ground state and metastable neutral Xe) are formed and aligned by electron impact in the same volume of space. Since both atoms with different states are aligned (not oriented), how one may obtain an observable effect due to spin-exchange collision is not so obvious as in the case of collisions between an oriented atom and an unoriented atom. By a rather qualitative consideration, one can account for the phenomena reported here. First, by electron impact along the field direction, the states

$$|^3P_2, m_J=0, m_S=\pm\tfrac{1}{2}\rangle, \quad |^3P_2, m_J=1, m_S=+\tfrac{1}{2}\rangle,$$
$$|^3P_2, m_J=-1, m_S=-\tfrac{1}{2}\rangle, \quad |^2P_{3/2}, m_J=\tfrac{1}{2}, m_S=+\tfrac{1}{2}\rangle,$$
$$\text{and } |^2P_{3/2}, m_J=-\tfrac{1}{2}, m_S=-\tfrac{1}{2}\rangle$$

are preferentially produced. From production and through subsequent spin-exchange collisions, an equilibrium obtains among these states. The introduction of a radio frequency that redistributes the populations of either the $^3P_2$ or $^2P_{3/2}$ states affects the absorption of linearly polarized light by the $^3P_2$ state.

Using the Born-Oppenheimer approximation, Bethe showed that there is a preferential population of magnetic sublevels of the excited triplet state.[3] In particular, in case of ^1S-^3P excitation, at the threshold energy of excitation, the $M_L=0$ state is populated predominantly. Lamb pointed out that one would not expect the results for the threshold excitation to be in serious error for bombardments a few volts above the threshold.[4] That this is true has been verified for the excitation of the triplet ($^3P_1$) state of He and for the $^3P_2$ metastable states of Hg and Ne.[5] Thus, the possible magnetic sublevels of the $^3P_2$ state $M_J=0$ and ±1 are formed, since the ground state of Xe has $^1S_0$ configuration. Let us consider only the $M_J=0$ case in detail, since similar consideration will hold for the $M_J=\pm1$ states. Since XeI states show good j-j coupling, the $^3P_2$ state can be expressed as $|(s_1l_1)j_1(s_2l_2)j_2, J, M\rangle$, where $(s_1l_1)j_1$ corresponds to a $5^2P_{3/2}$ parent state and is coupled with an $(s_2l_2)j_2=6^2S_{1/2}$ electron, resulting in $^3P_2$ with $M_J=0$ with a restriction of $M_L=0$. Therefore, we have[6]

$$\begin{aligned}|(s_1l_1)j_1(s_2l_2)j_2, J, M\rangle &= |(s=\tfrac{1}{2}, l=1)\tfrac{3}{2}, (s=\tfrac{1}{2}, l=0)\tfrac{1}{2}, 2, 0\rangle \\ &= (1/\sqrt{2})\,|(s=\tfrac{1}{2}, l=1)m_s=\tfrac{1}{2}, m_l=0\rangle\,|(s=\tfrac{1}{2}, l=0)m_s=-\tfrac{1}{2}\rangle \\ &\quad+(1/\sqrt{2})\,|(s=\tfrac{1}{2}, l=1)m_s=-\tfrac{1}{2}, m_l=0\rangle\,|(s=\tfrac{1}{2}, l=0)m_s=+\tfrac{1}{2}\rangle. \qquad (1)\end{aligned}$$

This means that in the state with $M_J=0$, there are two possible substates, $m_S=\frac{1}{2}$ and $m_S=-\frac{1}{2}$. Similarly, if we assume that $\Delta M_L=0$ also holds true for the ionization process to produce the $^2P_{3/2}$ ground state of Xe^+, the $^2P_{3/2}$ state may be represented as

$$\begin{aligned}|^2P_{3/2}\rangle &= (1/\sqrt{2})\,|(s=\tfrac{1}{2}, l=1)m_s=\tfrac{1}{2}, m_l=0\rangle \\ &\quad+(1/\sqrt{2})\,|(s=\tfrac{1}{2}, l=1)m_s=-\tfrac{1}{2}, m_l=0\rangle; \qquad (2)\end{aligned}$$

i.e., $M_J=\frac{1}{2}$ and $-\frac{1}{2}$ states are produced such that $m_S=+\frac{1}{2}$ holds for $M_J=\frac{1}{2}$ and $m_S=-\frac{1}{2}$ holds for $M_J=-\frac{1}{2}$.

Since it is highly probable that the spin-singlet collision causes the spin exchange, the original $^3P_2, M_J=0$ state becomes, through spin exchange,

$$|^3P_2\rangle = a_1|0\rangle + a_2|1\rangle + a_3|-1\rangle, \qquad (3)$$

where

$$|1\rangle = |(s=\tfrac{1}{2}, l=1)m_s=\tfrac{1}{2}, m_l=0\rangle\,|(s=\tfrac{1}{2}, l=0)m_s=\tfrac{1}{2}\rangle$$

and

$$|-1\rangle = |(s=\tfrac{1}{2}, l=1)m_s=-\tfrac{1}{2}, m_l=0\rangle \times |(s=\tfrac{1}{2}, l=0)m_s=-\tfrac{1}{2}\rangle.$$

With the exception of the factor $(2)^{-1/2}$, $|0\rangle$ has been expressed in Eq. (1). Probability amplitudes a_1, a_2, and a_3 depend on the production and exchange rates. In any case, the state with $M_J=\pm1$ becomes populated through exchange collisions.

The $|^2P_{3/2}\rangle$ state remains unchanged as far as the spin direction is concerned, since in a spin-exchange collision the $m_S=+\frac{1}{2}$ state with $M_J=\frac{1}{2}$ becomes the $m_S=-\frac{1}{2}$ state with $M_J=-\frac{1}{2}$ and vice versa.

Next, if one applies an h_{rf} oscillating radiofrequency field perpendicular to the externally applied magnetic field at the Larmor frequency of the $^2P_{3/2}$ state, complete population mixing occurs among the $^2P_{3/2}$ magnetic sublevels. Under such conditions, there is a rather interesting population redistribution such that the $M_J=0$ of the $^3P_2$ state is more populated. If the $M_J=+1$ and -1 of $^3P_2$ are initially more populated, the $M_J=0$ state is more populated without the rf resonance of the $^2P_{3/2}$ state. Unless these two effects occur in exactly equal amounts, it is possible to observe the spin-exchange collision. But the $M_J=0$ state is initially more populated than $M_J=1$ and -1, as can be approximately estimated from Born-Oppenheimer theory by using calculations similar to those of Lamb.[4]

The experimental apparatus employed is quite similar to that used in our previous experiment on the rf paramagnetic resonance of the $^3P_2$ metastable state of neon.[7] Instead of Ne, we filled the electron-gun excitation tube with Xe at a pressure of about 5×10^{-4} mm of Hg. This diode-structure electron gun was operated in a space-charge neutralization condition.[8] Under this condition, as much as 10% of the ground-state atoms can be ionized, resulting in a large number of ionized and aligned $^2P_{3/2}$ states ($p \sim 10^{-5}$ mm of Hg). The metastable-state density is about 10^{-7} mm of Hg. Changes of the Xe^+ ($^2P_{3/2}$) ground state through spin-exchange collision with the Xe ($^3P_2$) state were observed by monitoring the change in linearly polarized $\lambda=8409$ Å ($^3P_2$-$^3S_1$) resonance absorption by the Xe ($^3P_2$) state. The light propagates perpendicular to the direction of the electron beam. An applied external magnetic field can be either parallel or perpendicular to the electron-beam direction. Both orientations give about the same

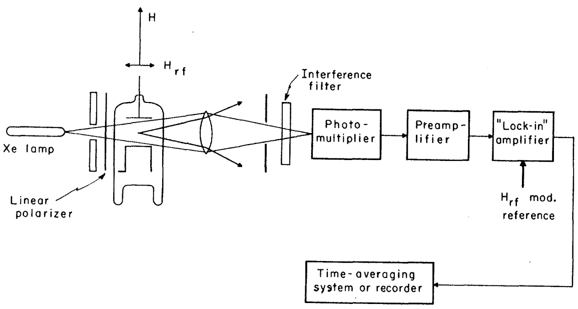

FIG. 1. Schematic drawing of apparatus. All electronic apparatus used is commercially available.

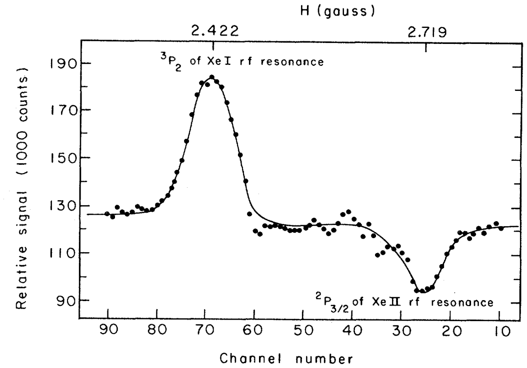

FIG. 2. Radiofrequency resonances of the $^3P_2$ and $^2P_{3/2}$ states with and externally applied magnetic field parallel to the electron-beam direction.

signal-to-noise ratio. Figure 1 is a schematic diagram of the apparatus.

Figure 2 shows the optical absorption of the cell as a function of the applied magnetic field. The signals shown are correlated with resonances in the $^3P_2$ and $^2P_{3/2}$ states of the even Xe isotopes. In addition, we observed resonances of Xe^{129} and Xe^{131} with $\sim\frac{1}{5}$ the signal strength. Normally the electron gun was operated at as much as 1 to 2 V below the ionization potential. We believe that in this case, the $^2P_{3/2}$ state is produced by a two-step process; i.e., the metastable state, $^3P_2$ as well as $^1P_0$, first produced by electronic impact, is ionized by a second electronic impact. However, more well-controlled experiments are obviously necessary to understand the mechanisms involved. In addition, it is interesting to note that the spin relaxation time of the Xe ($^1P_2$) state is about $\frac{1}{2}$ that of the Hg ($^3P_2$) state under almost identical experimental conditions.

By means of an extremely simple apparatus, we found that rf spectroscopy of the ionic ground state of Xe^+ having $^2P_{3/2}$ configuration is possible by using spin-exchange collisions with the $^3P_2$ metastable state as a detector. Since our experimental setup is sensitive to only diagonal density-matrix elements, i.e., on the average population of the magnetic substates, it was only possible to observe the rf resonance of ^2P state with $J=\frac{3}{2}$. However, Series has shown that by observing transfer of coherence, the spin-exchange effect can be seen even for $J=\frac{1}{2}$ by setting the rf frequency to correspond to the difference of the Larmor precessional frequencies of two different states.[9] Extension to hfs measurement of the ion is obvious and will be attempted. We are presently working on a Series-type experiment to look for the transfer of coherence through spin-exchange collisions, as well as on the hfs of Xe^+ ($^2P_{3/2}$) in Xe^{129} and Xe^{131}. It seems that this method should be easily applicable to other noble-gas atoms.

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