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Zeeman Effect in the Two-Photon 3S-5S Transition in Sodium Vapor*

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The technique of two-photon absorption without Doppler broadening has made possible the observation of the Zeeman splitting of a transition between two ^2S states. Selection rules are discussed. The observed pattern of hyperfine components is narrower in very high fields than in zero field, in agreement with the theory.

Several authors[1-3] have recently reported the observation of two-photon absorption resonances of the Na atom with high resolution by means of narrow-band tunable dye lasers. The Doppler broadening in the vapor is eliminated by absorbing one photon from each of two light beams moving in opposite directions.[4,5] In this note, we report the effect of a magnetic field on the two-photon resonant transition between the 3S and 5S levels of the Na^{23} atom. The features of the Zeeman effect in this case are novel and rather different from the features occurring in one-photon transitions.

Both the 3S ground state and the 5S excited state have a total angular momentum $\vec{F}=\vec{I}+\vec{S}$. The nuclear spin of Na^{23} is $I=\frac{3}{2}$; $S=\frac{1}{2}$ and consequently $F=2$ or 1. The spin Hamiltonian for both the initial and final S states is given by

$$\mathcal{H}=A\vec{I}\cdot\vec{S}+g\beta\vec{S}\cdot\vec{H}_0+g_I\beta\vec{I}\cdot\vec{H}_0. \quad (1)$$

The contact hyperfine interaction constants for the 3S state and the 5S state are given by $A_{3S}=886$ MHz and $A_{5S}=78$ MHz, respectively. The nuclear and electron g factors are the same in both orbitals. The energy levels of both hyperfine multiplets are given by the Breit-Rabi formula, and the corresponding spin states can be found. It will be convenient to label the eight spin states of both the initial and final orbitals by $|m_I, m_S\rangle$.

The selection rule on the orbital magnetic quantum number remains strictly $\Delta m_L=0$, since $L=0$ in both S states. Therefore, circularly polarized light, pure σ^- or pure σ^+ radiation, can never induce this two-photon transition, regardless of the magnitude or direction of the external magnetic field. Furthermore the Hamiltonian of the atom plus its interaction with the radiation field commutes with both $\vec{I}$ and $\vec{S}$, provided that the effect of the spin-orbit coupling $\lambda\vec{L}\cdot\vec{S}$ in the intermediate atomic p state is negligible.[1,5] If terms on the order of $\lambda/\hbar(\omega_{3p}-\omega_{3s}-\omega)$ may be ignored, the selection rules in low magnetic field are $\Delta F=0$, $\Delta m_F=0$. Since the Landé g factors are exactly the same for the initial and final states, there is no variation in the frequency of the allowed transitions in weak fields, satisfying the condition $g\beta H_0<A_{5S}$. We retain a doublet with a separation $2\Delta\nu=2h^{-1}(A_{3S}-A_{5S})$ with intensity ratio 5:3, as observed in zero magnetic field. The factor 2 on the left-hand side is inserted so that $\Delta\nu$ corresponds to the frequency variation in the light beams participating in the two-photon transition.

In very high fields, $g\beta H_0\gg A_{3S}$, the Zeeman energy of the $|m_I, m_S\rangle$ state is given by $Am_Im_S+g_I\beta H_0m_S+g_I\beta H_0m_I$. The selection rule in this case is $\Delta m_I=0$, $\Delta m_S=0$. The difference in fre-

quency between the eight allowed transitions $|m_I, m_S\rangle \rightarrow |m_I, m_S\rangle$ is given by $(A_{3S} - A_{5S})m_I m_S$. The high-field pattern consists therefore of four components of equal intensity. The frequency separation between the outer components is $2\Delta\nu = \frac{3}{2}h^{-1}(A_{3S} - A_{5S})$, which is smaller than the zero-field separation, and independent of the magnitude of H_0 in the high-field limit.

For intermediate field strengths the spin Hamiltonian can be factorized in two-by-two matrices (Breit-Rabi) and the eigenstates are linear combinations $C_+|m_I, +\frac{1}{2}\rangle + C_-|m_I+1, -\frac{1}{2}\rangle$. Since the spin decoupling in the $3S$ ground state occurs at a different field strength ($A_{3S}/g\beta \sim 600$ G) from that in the $5S$ state ($A_{5S}/g\beta \sim 50$ G), the spin wave functions of initial and final states are not identical. The appropriate projections of the initial spin state onto the final spin states must be taken. This leads, in general, to two transition frequencies for each initial spin state, except for the $|+\frac{3}{2}, +\frac{1}{2}\rangle$ and $|-\frac{3}{2}, -\frac{1}{2}\rangle$ states. For any field strength, these always contribute to a single frequency line at the same position as the $F = 2 \rightarrow 2$ component in zero field. In intermediate fields there are, in general, thirteen components. The predicted patterns are shown by vertical bars in Fig. 1, for a field of 570 G and for a field of 5400 G.

These features have been verified experimentally. The experimental apparatus has been described elsewhere.[1] A collimated beam, consisting of the 4-nsec pulses with 1 mm diameter, traversed the Na vapor cell. The power flux density was varied from 6 to 300 kW/cm^2. The density in the Na vapor cell was varied from 10^9 to 10^{12} atoms/cm^3. It was verified that the observed fluorescent signal at the $4P$-$3S$ transition, following the two-photon absorption, was proportional to the sodium density, and to the product of the intensities of the forward and backward beams which had opposite circular polarizations. The zero-field doublet splitting is recorded in the top of Fig. 1. The improved signal-to-noise ratio has led to an improved determination of the splitting $\Delta\nu = h^{-1}(A_{3S} - A_{5S}) = 808 \pm 5$ MHz, which leads to a value $A_{5S} = 78 \pm 5$ MHz. This value, which supersedes that reported in Ref. 1, is in excellent agreement with theoretical predictions.[6]

It was verified that the forward beam alone, if circularly polarized, never induces two-photon transitions for any direction of the magnetic field. The observed Zeeman patterns are independent of the direction of the magnetic field with respect to the light beam propagation, also in

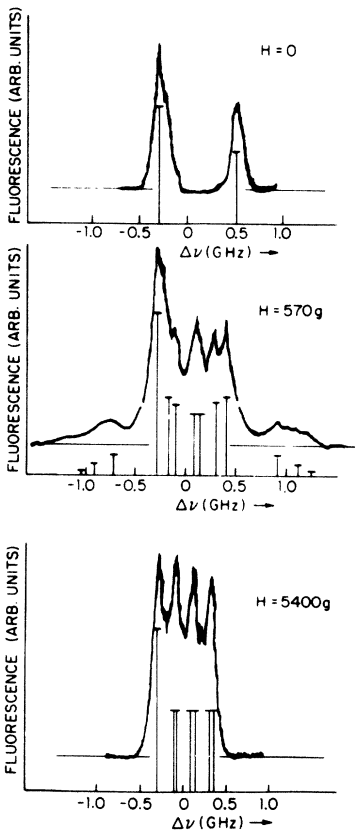

FIG. 1. Hyperfine splitting of the two-photon $3S$-$5S$ transition in the Na^{23} atom in zero magnetic field, and in magnetic fields of 570 and 5400 G, respectively. The vertical lines indicate the theoretically calculated line positions and their relative strengths. The experimental traces record the observed fluorescence intensity at 330 nm ($4P$-$3S$ transition) following the two-photon absorption process.

agreement with the theory. At the bottom of Fig. 1 the recorded pattern in a relatively high field, $H_0 = 5400$ G, is shown. The field homogeneity is not important in this case, as the pattern approaches the field-independent form, consisting of four equidistant lines of equal intensity.

In the middle of Fig. 1 the recorded pattern in an intermediate field, $H_0 = 570 \pm 10$ G, is reproduced. For comparison, the theoretically predicted line positions and relative strengths are shown by vertical bars. There is good agreement between theory and experiment. Note the weak satellites in the wings, corresponding to transitions $\Delta m_S = -\Delta m_I = \pm 1$, forbidden in the high-field limit. In order to record these wings, a Fabry-Perot interferometer with a larger free

spectral range, but with a concomitant lower resolution, had to be used. The central portion of the spectrum was recorded by tilting a high-resolution Fabry-Perot with a 5-cm plate spacing. The experimental resolution, corresponding to a full width at half-maximum of 150 MHz, is largely determined by the short pulse duration of 4×10^{-9} sec, in accordance with the uncertainty principle. Unfortunately this resolution is not sufficient to resolve all of the individual components.

In this context it should be noted that the nitrogen-laser–pumped dye laser available to us was designed for high peak power output and intended for the investigation of nonlinear optical properties of crystals and liquids. Hänsch has shown that much higher resolution of about 10 MHz can be obtained with cw dye lasers. The nonlinearity of many two-photon transitions in alkali vapors is so large that a focused beam with 0.1–1-W cw power is sufficient for their observation. Our limited resolution was, however, sufficient to obtain confirmation of the rather striking selection rules and Zeeman splittings governing two-photon transitions between one-electron ^2S states.

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Raman Scattering from Vibronic Levels of a Jahn-Teller–Distorted Complex*

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Raman-scattering transitions have been observed between the hindered rotational states of the Jahn-Teller–distorted ^2E_g ground state of Cu^{2+} in CaO. Impurity-induced scattering from the CaO vibrations was also observed. The results are in good agreement with the cluster model for the low-lying vibronic states.

The vibronic energy levels of Jahn-Teller–distorted impurity complexes in solids have been the subject of many theoretical efforts over the previous two decades. However, no unambiguous and extensive experimental information has been obtained in this area because of the lack of a spectroscopic technique which was not limited severely by frequency range, selection rules, or complications resulting from extraneous spectral information not related to the Jahn-Teller distortion. We report the first observation of Raman scattering from both the hindered rotational levels and the impurity-perturbed lattice vibrations associated with a Jahn-Teller distortion. The experiments were performed on the ^2E_g electronic ground state of the Cu^{2+} ion at a Ca site in CaO. The substantial intensity of the spectra, particularly for the transitions between the rotational levels, suggests that this will be a powerful and widely applicable method for studying the Jahn-Teller effect.

The experiments were performed using the 5154-Å argon laser line and conventional photon-counting detection techniques. An I_2 filter[1] was employed for the work within 200 cm^{-1} of the laser line to reduce the intensity of the Rayleigh-scattered laser light. The samples were grown at Oak Ridge National Laboratory by an arc-fusion method and had varying Cu^{2+} concentrations from sample to sample as well as over the volume of each sample. Spectra were obtained for polarization geometries for which the E_g, $E_g + A_{1g}$, and T_{2g} scattering symmetries were observed.

The E_g spectrum at 4.2°K for the 0–200-cm^{-1} region is shown in Fig. 1. Four sharp lines are observed at 4, 26, 34, and 38.5 cm^{-1}. In addition, three broader and weaker peaks appear at