HYPERFINE STRUCTURE AND PARAMAGNETIC PROPERTIES OF EXCITED STATES OF XENON STUDIED WITH A GAS LASER*

H. R. Schlossberg and A. Javan
Physics Department, Massachusetts Institute of Technology, Cambridge, Massachusetts
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In a recent paper[1] we have presented a detailed theoretical analysis of an effect[2] which is of considerable spectroscopic value in the determination of hyperfine structure and paramagnetic properties of highly excited atomic levels. This Letter gives the first application of this effect to the determination of hyperfine structure; specifically, that of a highly excited electronic state of ^{129}Xe. There exist numerous other excited states with hyperfine structure in xenon and other atoms which may now be studied similarly. We also report measurements of the g values of several excited states of Xe, together with estimates of their radiative lifetimes.

The experiments presented here involve the saturation behavior of a Doppler-broadened transition consisting of levels with closely spaced structural components.[1] The transition is considered to interact with a radiation field consisting of several optical frequencies lying within the Doppler width. The frequency spacings of this field are assumed to be known. Consider the case that the level structure is tuned —e.g., by application of a dc magnetic field. Then, the coupling of the optical field with the atomic transition appreciably changes[3] whenever the spacing of an appropriate pair of upper (or lower) energy-level components becomes equal to one of the frequency spacings of the applied field. This change has a resonant form with a width determined only by the radiative lifetime of the two corresponding level components. When the level structure arises from hyperfine interaction, appropriate pairs are those belonging to $|\Delta M_F| = 0$ or 2 (where M_F is the projection of the total angular momentum F onto the magnetic field. Details of this effect have been already described elsewhere. Here we emphasize the use of the effect as an inherently simple experimental tool and its application to measurements of hfs.

In the present experiments we have used a multimode, linearly polarized, Brewster-angle gas laser. The resonator was close to a confocal configuration. An iris diaphragm was used near each mirror to limit the laser to axial-mode operation. In this case the laser radiation contains a number of frequencies whose separations are closely given by integer multiples of $\nu_m = c/2L$, where L is the length of the resonator.[4] An axial magnetic field was applied to the laser tube by means of a solenoid. The above-discussed coupling effect appeared in the form of a resonant change in the output power[1] whenever the spacing of the appropriate Zeeman sublevels became equal to the frequency separation of two oscillating modes. For purposes of narrow banding, the magnetic field was modulated by a small amount at an audio frequency. This allowed the use of a phase-sensitive detector. The dc output of the phase-sensitive detector was fed to the y axis of an x-y recorder, with the x axis driven proportionally to the solenoid current. Thus, the curves obtained have the form of a derivative of the resonance signal.

The tracings in Fig. 1 are samples of the experimental results for the 3.37-μ line ($5d[2\frac{1}{2}]_2^0$ to $6p[1\frac{1}{2}]_1$ in Racah notation) in Xe. The resonances in the figure are all due to splittings of the upper laser level. This is because of the very narrow radiative width (hundreds of kilocycles per second) of this level.[5] The lower level is much broader and resonances due to its structure are not seen under the present experimental conditions. This fact considerably simplifies the interpretation of the observed resonances.

The signals in the figure can be explained readily in terms of the energy-level diagrams of Fig. 2. First consider the various even isotopes of naturally occurring Xe which do not possess hyperfine structure. Their energy-level diagram is that of Fig. 2(a) which is the usual linear Zeeman effect. In this case the "selection rule" reduces to $|\Delta M_J| = 2$. The angular momentum of the level under consideration is $J = 2$. Note that for this level the selection rule allows three pairs of Zeeman sublevels with the same spacing. Accordingly the resonance due to each pair appears at the same value of magnetic field and their three signals are superimposed. The condition for

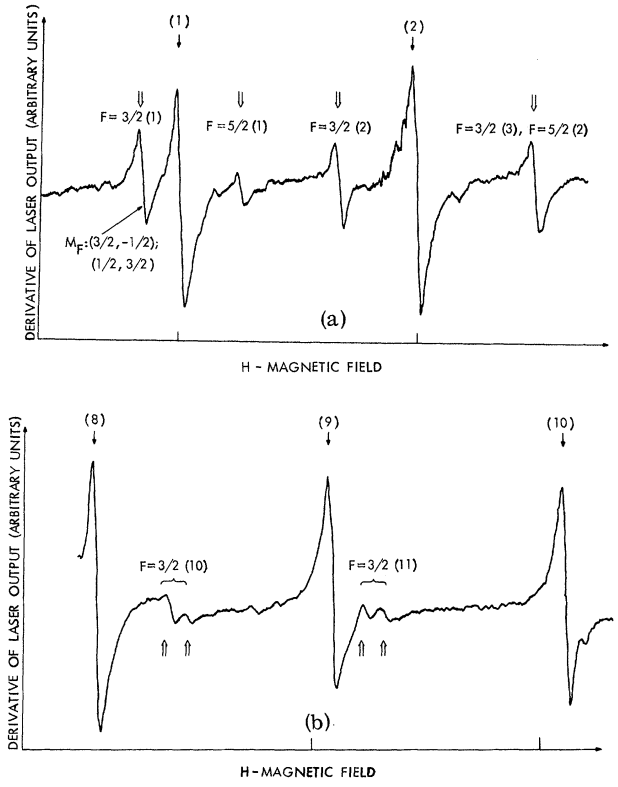

FIG. 1. Resonances of 3.37-μ transition of Xe. The orders of resonances appear in parentheses. Resonances indicated by a single arrow are due to even isotope of Xe and those indicated by double arrows belong to ^{129}Xe.

resonant coupling is $2g_J\beta H = n\nu_m$. Here g_J is the level g factor, β is the Bohr magneton, and n is an integer equal to the number of Fabry-Perot spacings, ν_m, between the pair of oscillating modes responsible for a specific resonance. We will henceforth refer to n as the order of the resonance signal.

The signals of Fig. 1 labeled with single arrows are due to the splittings of the even-isotope sublevels. The order of each resonance is indicated in parentheses.

Now consider the odd isotope ^{129}Xe which has a nuclear spin $\frac{1}{2}$ and occurs in sizable abundance in a normal xenon sample. In this case the hfs of the level under consideration has two components corresponding to $F = \frac{3}{2}$ and $F = \frac{5}{2}$. The energy-level pattern of this isotope is given in Fig. 2(b). The resonances of Fig. 1 indicated by double arrows belong to this isotope and correspond to $|\Delta M_F| = 2$. At a low value of the magnetic field the Zeeman splitting of each hyperfine component is linear, corresponding to g values $g_{3/2} = (6/5)g_J$, $g_{5/2} = (\frac{4}{5})g_J$.[6]

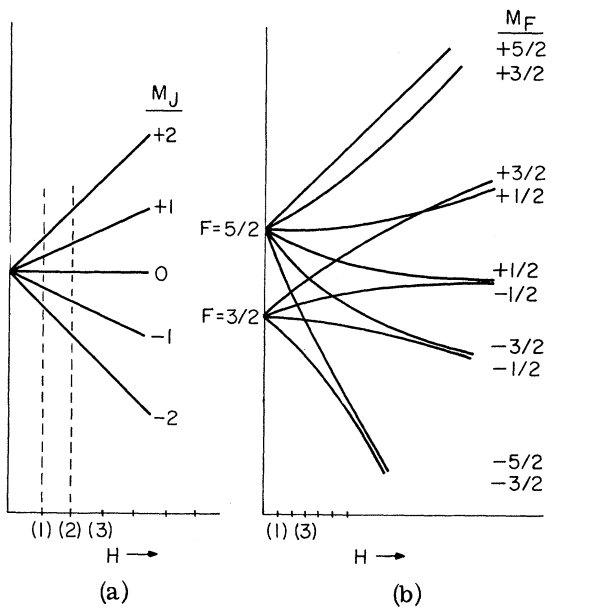

FIG. 2. Energy-level diagrams. The scale in (a) is distorted and differs from (b).

Because of this linear dependence, low-order resonances are due to a superposition of the effects of several $|\Delta M_F| = 2$ pairs, as in the case, described above, of the even isotopes. At a large value of the field, however, the linear dependence no longer holds. Accordingly, the above-mentioned $|\Delta M_F| = 2$ resonances, when obtained at a high order, are no longer superimposed. In fact, by comparing the successive orders of a resonance corresponding to a given F value, details of their gradual splittings may be followed with ease.[7] In effect we sample successively the energy intervals of the Breit-Rabi pattern at many discrete frequencies. This fact makes the identification of observed resonances particularly simple.

From the departure from a linear Zeeman effect illustrated by the doublets in Fig. 1, we have calculated the zero-magnetic-field hyperfine interaction constant, a (where the interaction Hamiltonian is $a\vec{I}\cdot\vec{J}$), to be 890 MHz with a most probable error of about 8%. The size of this error may be reduced appreciably by operating at larger values of the magnetic field where the departure from a linear Zeeman effect is more pronounced. There has been only one previous measurement[8] of the hfs of ^{129}Xe. This measurement was done on a metastable electronic state, $6s[1\frac{1}{2}]_2$, using a molecular-beam technique. The coupling constant

Table I. Measured g values and radiative linewidths of excited Xe levels.

Level	Transition (μ)	g Value	Width (MHz)
$5d[2\frac{1}{2}]_2^0$	3.37	0.929	0.60
$5d[3\frac{1}{2}]_3^0$	3.50	1.070	1.0
$5d[3\frac{1}{2}]_4^0$	5.57	1.254	0.85
$5d[1\frac{1}{2}]_2^0$	3.27	1.376[a]	0.35

[a]Known previously.

a in that case is a factor of about 3 greater.

The g values of several other $5d$ electronic levels in Xe are given in Table I. These values were measured by studying the laser transition listed in column 2 and are accurate to within 0.1%. The spacing between the even-isotope resonance signals of each transition was compared with that of the 3.27-μ transition. The g value of the upper level ($5d[1\frac{1}{2}]_2^0$) of this transition is known.[9] This made detailed calibration of the magnetic field unnecessary.

The radiative linewidths of the levels are also given in the table. They are obtained from the measured resonance linewidths and the calculated g values. These values are to be taken as rough estimates due to the presence of some inhomogeneity in our magnetic field. They are in good qualitative agreement with an existing theoretical estimate[10] of the radiative lifetime of these levels.

Here we should point out that the dependence of the width of a resonance signal only on the radiative lifetime of the two upper-level components (and not on the lifetime of the common lower level of the optical transitions) is an interesting and important feature which was discussed theoretically in Ref. 1. The narrow widths of the resonances obtained in the present experiments offer a direct proof of this fact.

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[1]H. R. Schlossberg and A. Javan, Phys. Rev. 150, 267 (1966).

[2]M. S. Feld, J. H. Parks, H. R. Schlossberg, and A. Javan, in Physics of Quantum Electronics, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (Mcgraw-Hill Book Company, Inc., New York, 1965), p. 567.

[3]The mechanism of the effect is a delicate interplay of saturation of level populations and double-quantum Raman coupling between various frequency components of the radiation field. (See Ref. 1.)

[4]Most of the present work was done with $\nu_m = 57.1$ MHz, corresponding to a mirror spacing of 2.65 cm. This could easily be varied, however, since the laser system was an external mirror; and several other spacings were also used. The values of ν_m were also checked by observing the beat frequency between two modes with a radio receiver attached to a fast infrared photodetector.

[5]All of the Xe laser oscillators in these experiments utilized pure Xe with pressures of a few mTorr. Thus, collision broadening effects were minimal.

[6]The operating values of magnetic field were too small to observe the sign of the coupling constant a. This sign may be determined, as usual, when the coupling of the nuclear moment with the field is appreciable. In Fig. 2, this sign is assumed to be positive for purposes of discussion.

[7]In our work we have observed as many as 21 orders of the resonance signal due to the even isotopes, and 16 due to ^{129}Xe.

[8]W. L. Faust and M. N. McDermott, Phys. Rev. 123, 198 (1961).

[9]C. E. Moore, Atomic Energy Levels, National Bureau of Standards Circular No. 467 (U. S. Government Printing Office, Washington, D. C., 1958), Vol. III.

[10]F. Horrigan, unpublished.