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OBSERVATION OF HYPERFINE LEVEL CROSSING IN STIMULATED EMISSION*

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A level-crossing effect in stimulated emission is used to determine precisely the hfs in an excited electronic state of Xe^{129}. The effect arises from a resonant change in nonlinear polarization induced by a monochromatic field as appropriate tunable level pairs are made to cross.

Several aspects of the nonlinear coupling of an intense monochromatic field with a Doppler-broadened transition having closely spaced structure have been discussed previously.[1-3] Experimental techniques based on these effects have yielded high-resolution information on the line structure, e.g., hfs[4] and isotope shift.[5] This Letter reports the observation of another effect of this class which may be defined as a stimulated level-crossing effect. The terminology is used to distinguish this from a similar effect observed in spontaneous emission from atoms pumped by a broad-band source.[6] In the stimulated level-crossing effect, one is concerned with the transmission of intense monochromatic light through a gas possessing a Doppler-broadened resonance with closely spaced structure; the effect manifests itself as a nonlinear change in the induced polarization as appropriate tunable atomic levels are made to overlap within their natural widths. This results in a change in the transmission coefficient, i.e., gain or attenuation of the incident radiation when the level crossing occurs.

A host of atomic and molecular laser transitions exist where this effect makes possible their high-resolution study. This Letter presents an application to precise determination of the hfs in one of the laser transitions in Xe.[7] Emphasis is given to a number of experimental procedures which facilitate observation of the effect.

The size of the level-crossing signal is proportional to the fourth power of the electric field strength; consequently, it is advantageous to utilize the high fields produced within a laser resonator. In this case, the laser medium serves two functions. First, it provides the saturating optical frequency field. Second, changes in laser polarization due to level crossings within the medium result in a resonant change in the output power.

In this experiment the hfs of the $5d(\frac{5}{2})_2{}^\circ$ state in Xe^{129} was determined by observing level crossings as the 3.37-μ laser transition $5d(\frac{5}{2})_2{}^\circ \to 6p(\frac{3}{2})_1{}^\circ$ was tuned via an axial magnetic field. The nuclear spin of Xe^{129} is $\frac{1}{2}$; therefore, at zero magnetic field there are two hyperfine components corresponding to total angular momentum of $\frac{5}{2}$ and $\frac{3}{2}$. A sketch of the energy level versus magnetic field is shown in Fig. 1.

The level-crossing signal may only be observed in cases where the optical transitions originating from the crossing levels terminate on a common lower level. Because of the relative orientation of the laser field and the magnet-

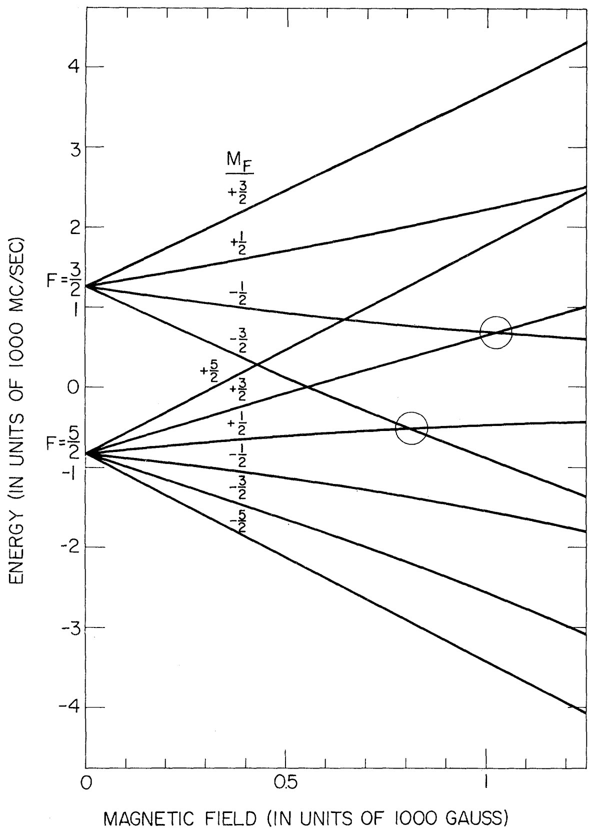

FIG. 1. Plot of hyperfine energy levels of the upper laser states of Xe^{129} as a function of magnetic field. The hyperfine interaction is given by $\hbar a \vec{I}\cdot\vec{J}$ and a is assumed to be negative as dictated by theoretical considerations of the electronic states involved, $5d(\frac{5}{2})_2°$. The observed level crossings are indicated by circles.

ic field, matrix elements exist only between states for which $\Delta F = 0, \pm 1$ and $\Delta m_F = \pm 1$. Three level crossings are therefore expected as the magnetic field is varied, the first of which should appear in the neighborhood of 800 G.

At the low gas pressure used where collision effects are negligible, the level-crossing line shape is essentially a Lorentzian whose width is given by the average of the radiative widths of the two crossing levels (0.5 Mc/sec in Xe). This holds in the case where the Doppler width is considerably larger than the radiative width of the optical transition. Note that the latter width is determined by the average radiative width of the upper and the lower level which in this case is approximately 10 MHz (the Doppler width of the transition is 200 MHz). Additional discussions of the line shape are given below.

In our first attempt, a solenoid provided a homogeneous dc magnetic field over the entire length (1 m) of the Xe discharge tube. An exhaustive search, however, failed to reveal the level-crossing signal.

The Xe sample used in this experiment consisted of a mixture of various isotopes composed of 60% Xe^{129} and 40% even isotopes Xe^{128}-Xe^{136} and Xe^{131}. It must be noted that in fields as high as 800 G the 3.37-μ line consists of a multitude of transitions arising from magnetic sublevels of both the even and the odd isotopes some of which are well resolved with respect to their Doppler widths. In such a case, the laser oscillation does not in general occur on all components of the line. This behavior arises from well-known nonlinear coupling between various laser transitions. On the other hand, a level-crossing signal cannot be obtained unless laser oscillation is sustained at frequencies within the overlapping Doppler widths of the transitions associated with the crossing levels. In our initial experiments this condition appears not to have been satisfied.

The above difficulty was overcome by making the following modifications in the experimental apparatus. The discharge was subjected to two separate regions of magnetic field provided by a Helmholtz coil and a solenoid. The solenoid produced a homogeneous dc field over 80 cm of discharge thereby tuning the bulk of the laser medium and determining the frequency of various oscillating laser modes. The Helmholtz coil, placed at the end of the laser discharge tube, produced a magnetic field uniform to within ±0.1% over a spherical volume of 4-cm diam centered on the symmetry axis. This coil provided the necessary homogeneous field used in searching for the level-crossing signal. Let us designate by H_c the value of the field at which a level crossing occurs and denote by ν_c the center frequency of the crossing optical transitions at this field. Inspection of the Zeeman splittings of various optical transitions belonging to the several isotopic species of Xe shows that it is possible to tune the center frequencies of some Zeeman components of the 3.37-μ transition through ν_c. This occurs at several field values differing appreciably from H_c. To insure the presence of an oscillating laser mode at a frequency within the Doppler width of ν_c, the solenoid field was tuned initially in coarse steps of about 20 G (corresponding to a shift of various transition frequencies by an amount of the order of the Doppler width); at each step a search was made for the observation of the level crossing by continuously tuning the Helmholtz coil. For purposes of narrow banding, the magnetic field, produced by the

Helmholtz coil, 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 Helmholtz current. Thus the curves obtained have the form of a derivative of the resonance signal.

Proceeding in this way we have observed two stimulated level crossings in Xe^{129}. They occur for values of the Helmholtz field of 802.2 and 1015 G, respectively, each having a width of approximately 1 G. In both instances, the signal was observed for several values of the solenoid field; small variations of the solenoid field (tens of gauss) around these values produced only a change in the strength of the observed level-crossing signal.

Before proceeding with further discussions of the results, additional significant experimental details must be considered: As the Helmholtz field is continuously tuned, additional resonances, unrelated to the level-crossing signals, are observed. The origin of these signals is well known. They arise from laser multimoding and occur whenever two appropriate Zeeman components are separated by the difference in frequency between two simultaneously oscillating laser modes. It should be noted that the magnetic field at which these so-called mode crossings occur depend upon the frequency separation between adjacent laser modes, and hence on the length of the laser resonator. However, the level crossing is associated with each oscillating laser mode individually and occurs at a value of magnetic field independent of the frequency spacing between modes. Advantage was made of this property in identifying the level-crossing signals; a mechanical sweeping device was used to translate one of the laser mirrors sinusoidally with an amplitude of about 2 cm and with a period short compared with the averaging time of the detection system; this resulted in smearing of the mode-crossing resonances while the level-crossing signals were unaffected. Figure 2(a) is a tracing obtained with fixed resonator length; note the numerous mode-crossing signals.[4] Figure 2(b) is a tracing over the same region of field while the resonator length is periodically modulated; the mode-crossing resonances are entirely eliminated. Initially, however, a sweeping device was not employed. Rather, the mirror spacing was changed in discrete steps and a tracing was obtained by varying the magnetic field over the level-crossing region for each resonator length. Comparison of the tracings obtained in this way made possible the identification of the level-crossing resonance as the signal which appeared at the same value of the magnetic field irrespective of the resonator length.

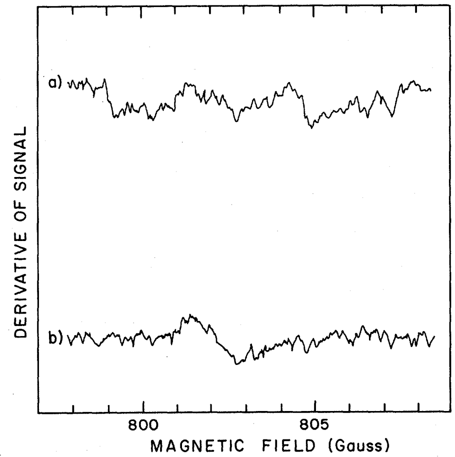

FIG. 2. Recorded tracings obtained as the magnetic field is swept through the region of a level crossing; (a) was obtained with a fixed resonator length, note the presence of mode-crossing signals; (b) shows the result of modulating the resonator length, the mode crossings are eliminated leaving only the level-crossing signal.

The magnetic field at which level crossings occur was determined with an NMR gaussmeter; magnetic field inhomogeneities limited the accuracy to 5 parts in 10^4. An order-of-magnitude improvement may be obtained with a better magnet. This observation was used to determine the size of the hyperfine interaction $\hbar a\vec{I}\cdot\vec{J}$ as $a = 829.5 \pm 0.8$ MHz.[8,9] This also agrees with a previous, less precise measurement made on the basis of a mode-crossing experiment in which the hyperfine constant was determined[4] by observing small departures from linear Zeeman splittings at magnetic field values below 200 G.

A preliminary theoretical analysis of the level-crossing effect in stimulated emission has been given in an earlier publication[10] in which emphasis is placed on describing the effect and estimating the size of the level-crossing signal. Information about the line shape may be obtained only through a more detailed analysis[11] in which the induced polarization is computed to third order in the electric field as a function of the upper level spacing. If the optical-frequency field is assumed to take the form of a traveling wave then the calculation is simplified and it is found that the line shape is that of a narrow Lorentzian (whose width is the average homogeneous width of the crossing levels) superimposed on a broad Gaussian response characterized by the Doppler width. The optical field within a laser resonator is of course a standing wave. In this case the calculations are somewhat lengthy; however, the results exhibit the simple behavior of the traveling wave field if the optical-frequency field lies on the side of the Doppler profiles of the two crossing transitions. When the optical-frequency field lies close to the line center, additional resonances, related to the Lamb dip, become important. These, however, are characterized by a width given by the average of the widths of the upper and lower levels; they are in general rather broad and consequently do not interfere with the observation of a narrow, simple Lorentzian level-crossing line shape.

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OPTICAL ORIENTATION OF THE $^3P_0$ GROUND STATE OF Pb^{207}

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The method of optical pumping of diamagnetic atoms with zero orbital angular momentum ($^1S_0$) has been extended to an atom with nonzero orbital angular momentum ($^3P_0$), yielding $|\mu_I|_{\text{uncorrected}} = 0.5726(6)\mu_N$ for the nuclear moment of Pb^{207}. The cross sections for disorientation by collisions with the inert gases, hydrogen, and nitrogen are all less than 10^{-20} cm^2.

The optical pumping of the $^3P_0$ ground state of Pb^{207} reported herein is the first nuclear resonance observed in free diamagnetic atoms with nonzero orbital angular momentum. This is a new extension of the technique of optically pumping diamagnetic atoms, heretofore restricted to states with zero orbital angular momentum, i.e., $^1S_0$ states.[1] The present measurement of the nuclear moment of Pb^{207} demonstrates the applicability of the technique to determining nuclear moments of $^3P_0$ atoms, free from chemical shifts and from the strong electron-nuclear interactions of paramagnetic states. The upper limits placed on the cross sections for collisional disorientation by various buffer gases are particularly interesting in that they are much smaller than the