Motional Stark Effect in High Magnetic Fields: A New Technique for Sub-Doppler Spectroscopy

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(Received 8 July 1977)

We have observed transitions between Rydberg states of He with sub-Doppler resolution, using a novel technique. Ordinary Doppler line shapes, observed at low magnetic fields, are completely altered at high fields by the quadratic Stark effect induced by velocity-dependent electric fields arising from atomic motion transverse to the magnetic field. The high-field line shape has an abrupt cutoff, corresponding to $v_\perp \simeq 0$ atoms, whose slope is primarily limited by the homogeneous linewidth.

The Doppler effect has long been a significant obstacle to high-resolution spectroscopy. Historically, efforts to overcome the Doppler effect have centered on two principal techniques: the use of molecular (atomic) beams and—particularly with the advent of lasers—the interaction, under suitable conditions, of molecules with more than one photon.[1] Besides these two direct approaches, double-resonance and anticrossing techniques have provided indirect means of obtaining sub-Doppler spectroscopic resolution.[2]

We report in this Letter a new technique, fundamentally different from any of the above, for obtaining sub-Doppler spectra. This technique exploits another velocity-dependent interaction in the molecular (atomic) Hamiltonian—that due to the electric field, $\vec{E}_s = c^{-1}\vec{v}\times\vec{B}$, seen by a molecule (atom) in its rest frame while moving through a magnetic field. If a particular atom has only a quadratic Stark effect, then all the atoms in the gas will have their energy shifted in the same direction by an amount proportional to $|\vec{E}_S|^2$, independent of the direction of their velocity. This has the effect of altering the normal Doppler profile by sharpening it on one side of the $v=0$ atomic line center, and broadening it on the other side. We have directly observed this effect in infrared magnetic-resonance transitions between Rydberg states of the He atom.

The experimental apparatus represents a modification of the equipment previously employed[3] for anticrossing experiments between levels of atoms and molecules differentially excited by an electron beam. As Fig. 1 shows, the prinicpal modification consists of the introduction of a CO_2-laser beam into the luminous sample region. The atomic energy levels are magnetically tuned to resonance with the fixed-frequency step-tunable, continuous-wave, CO_2 laser. The laser frequency is not directly measured. Rather, the laser is operated in a low-gain mode at the top of the gain curve, and the frequency is inferred to within a few MHz. Since Rydberg n, l states are differentially excited by electron impact, net pumping of atoms between these states occurs when they are Zeeman tuned to the laser frequency. The resonance condition is determined by monitoring the easily detected luminescence of optical photons (rather than the infrared photons which are harder to detect) emitted when the excited state decays radiatively. With the 140-kG NMR magnet that we are using, as many as ten different laser frequencies can resonate with the same set of levels at different fields. The magnetic field is measured with a broadband NMR probe. Field inhomogeneities and instabilities in our feedback-stabilized magnet are on the order of 10 ppm.

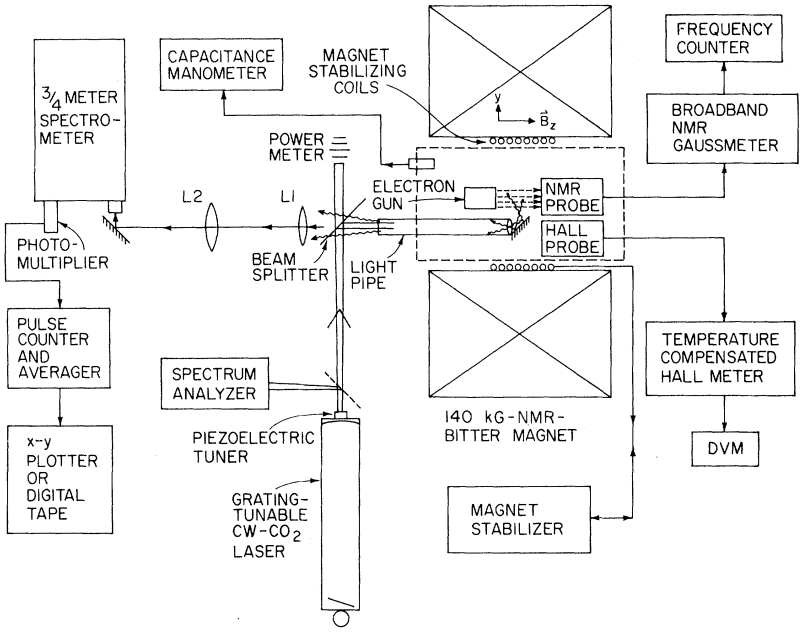

FIG. 1. Schematic diagram of experimental apparatus.

We have observed, among others, the $7^1S \rightarrow 9^1P$ transition in He. At low magnetic field (~2 kG) this line has a half width at half-maximum of 85 G = 119 MHz. This corresponds to the expected Doppler width at a temperature of ~150°C when one includes a small correction for the homogeneous linewidth. This temperature is slightly above ambient but quite reasonable as we know from other evidence that the whole sample region is somewhat heated by the ~50 W needed to operate the electron gun. At this level, the present work is analogous to several recent resonance experiments on Rydberg states of Na and He, and holds considerable promise for providing complementary information to these experiments. The experiments include work on Na by Ducas and Zimmerman[4] in the infrared and by Gallagher, Hill, and Edelstein[5] using microwaves. MacAdam and Wing[6] have earlier performed a series of microwave experiments on He itself.

A very important difference between the present experiments and the previous ones is our use of a large magnetic field. Figure 2(a) shows the same $7^1S \rightarrow 9^1P$ transition that we observed at 2 kG with an 85-G half-width, but now at a high magnetic field (and different laser frequency). The line shape is clearly different from a Doppler profile and, most importantly, the leading edge of the line shows a pronounced sharpening. This effect is seen even more dramatically in Fig. 2(b) for a different laser frequency at higher magnetic field where, on the enlarged scale, the line shape approaches a step function.

The explanation of the cutoff line shapes can be found by considering the Stark effect produced by the electric field, $\vec{E}_S$, that the atom sees in its rest frame. For an atom of given velocity $\vec{v}$, the resonance condition in either a field- or frequency-swept experiment can be written as

$$\Delta\omega = \omega - \omega_0 = g_e \mu_0 (B - B_0) = \frac{v_y}{c}\omega_0 + \frac{(v_x^2 + v_y^2)}{c^2}\alpha B^2, \quad (1)$$

where g_e is the effective g factor as determined by the linear Zeeman effect and the parameter

$$\alpha = \frac{1}{\hbar}\left(\sum_n \frac{(\mu_\perp^n)^2}{\Delta E_n} - \sum_{n'} \frac{(\mu_\perp^{n'})^2}{\Delta E_{n'}}\right)$$

gives the Stark coupling for the levels involved in the transition, where $\mu_\perp^n$ is the appropriate matrix element of the component of the atomic-dipole-moment operator perpendicular to $\vec{B}$, and ΔE_n the perturbation energy denominator. The first term on the right-hand side of Eq. (1) describes the usual Doppler shift and the second term is the quadratic Stark shift due to the effective electric field. These terms have been written down for a coordinate system appropriate to our experiment (Fig. 1). The laser beam is along y and $\vec{B}$ is normal to the x-y plane.

One can readily see from Eq. (1) that not all

values of v_x will be allowed to participate in the resonance (as long as v_x is required to be real). In fact, one obtains from Eq. (1)

$$v_x = (a^2 - u^2)^{1/2}, \tag{2}$$

with

$$a^2 = \frac{c^2}{\alpha B^2}\left[\Delta\omega + \frac{\omega_0^2}{4\alpha B^2}\right] \tag{3}$$

and $u = v_y + \omega_0 c/2\alpha B^2$. Since a^2 must always be a positive quantity, we can see from Eq. (3) that $\Delta\omega$ also has a cutoff value beyond which the resonance must disappear. The cutoff will occur at a maximum or minimum $\Delta\omega$, depending on the sign of α.

To obtain the line-shape function at the frequencies allowed by Eq. (1), $\alpha^{-1}[\Delta\omega + (\omega_0^2/4\alpha B^2)] > 0$, one must find the number of molecules with allowed values of v_x, v_y, and v_z. This is done by integrating a Maxwell-Boltzmann distribution

$$g(v_x, v_y, v_z) = (1/\pi v_0^2)^{3/2} \exp[-(v_x^2 + v_y^2 + v_z^2)/v_0^2] \tag{4}$$

over all v_z and all v_y allowed by Eqs. (1) and (2). In Eq. (4) we have assumed a thermal velocity distribution with $v_0^2 = 2kT/M$. After appropriate substitutions of Eq. (1) into Eq. (4) and a change of variables, one obtains the expression

$$f(\omega) = \frac{c^2}{2v_0^2\alpha B^2\pi} \exp\{-(c^2/v_0^2\alpha B^2)[\Delta\omega + (\omega_0^2/2\alpha B^2)]\} \int_{-a}^{a} \frac{\exp(\omega_0 cu/v_0^2\alpha B^2)}{(a^2 - u^2)^{1/2}}\, du. \tag{5}$$

The limits on the integral come from the velocity cutoff implied by Eq. (2) as discussed above. The integral in Eq. (5) is a modified Bessel function and the final line shape is given by

$$f(\omega) = \begin{cases} \dfrac{c^2}{2v_0^2\alpha B^2} \exp\left[-\dfrac{c^2}{v_0^2\alpha B^2}\left\{\Delta\omega + \dfrac{\omega_0^2}{2\alpha B^2}\right\}\right] I_0(\beta) & \text{for } \dfrac{1}{\alpha}\left[\Delta\omega + \dfrac{\omega_0^2}{4\alpha B^2}\right] > 0; \\ 0 & \text{for } \dfrac{1}{\alpha}\left[\Delta\omega + \dfrac{\omega_0^2}{4\alpha B^2}\right] < 0; \end{cases} \tag{6}$$

where $\beta = \omega_0 ca/\alpha v_0^2 B^2$. For the field-swept case, Eq. (6) can be trivially modified by substituting for $\Delta\omega$ from Eq. (1).

Equation (6) becomes somewhat more physically transparent when one considers the limiting case of a large motional Stark effect (i.e., when $\gamma = v_0\alpha B^2/\omega c\sqrt{\ln 2}$, the ratio of the motional Stark effect to the undisturbed Doppler width, $\Delta\omega_D = \omega v_0\sqrt{\ln 2}/c$ is large; $\gamma \gtrsim 10$). In this case the cutoff value of $\Delta\omega \approx 0$, and $I_0(\beta) \approx 1$, since $\beta \ll 1$. The resultant line shape is, therefore, zero on one side of ω_0 (or B_0), rises discontinuously, and then decays as a slow exponential.

The final interpretation of an experimental line shape must also include a convolution of Eq. (6)

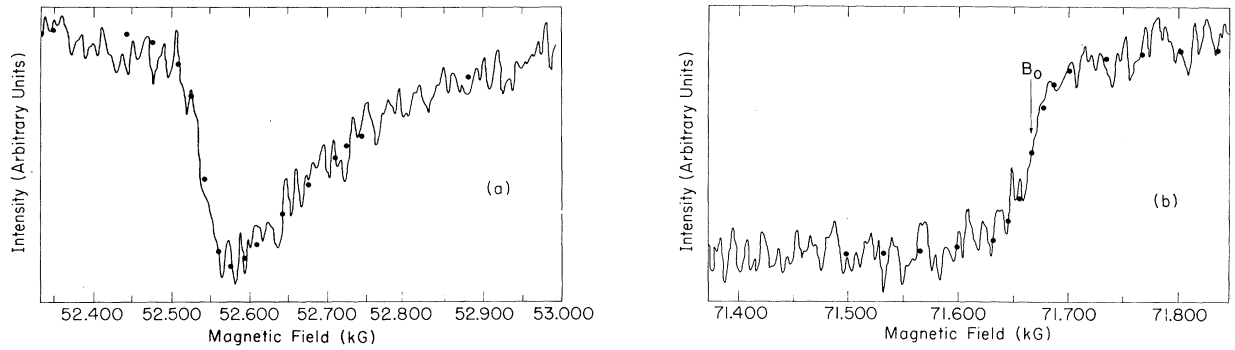

FIG. 2. Laser magnetic-resonance transitions between the $7^1S \rightarrow 9^1P$ He states: (a) $M_L = 0 \leftrightarrow M_L = -1$, via the $R24$ 10.6-μm laser line; (b) $M_L = 0 \leftrightarrow M_L = +1$, via the $R24$ 10.6-μm laser line. The 7^1S-2^1P emission intensity at 4024 Å was monitored. Typical experimental conditions include pressure, 10 mTorr; electron current, 1 mA, electron voltage, 100 V; laser power, 5 mW; and averaging time, 30 min. The dots are calculated points assuming $\Delta B = 14$ G. The line shape changes between (a) and (b) not only because of the increase in magnetic field but also because the magnitude and *sign* of α change.

with a velocity-independent shape inherent to the molecules and the experiment (e.g., homogeneous line). The convolution has the effect of softening the sharpness of the cutoff. In fact, the cutoff acquires a slope that is proportional to the linewidth of the velocity-independent line. The frequency difference between the center of the sloping cutoff and the actual atomic line center for a $v_\perp \simeq 0$ atom at high field is given by $\Delta\omega_D(4\gamma\sqrt{\ln 2})$.

We again consider the analytically simple case for which γ is sufficiently large. As discussed above, in this limit $f(B)$ or $f(\omega)$ near the cutoff can be well represented by a simple step function. Upon convolving this step function with a Lorentzian of half-width ΔB, centered on B_0, one obtains for the line shape

$$f(B) \propto \{\tfrac{1}{2} - \pi^{-1}\tan^{-1}[(B - B_0)/\Delta B]\}. \quad (7)$$

The solid dots in Fig. 2(b) are a fit with Eq. (7) [$\gamma \approx 12$] and indicate good agreement with experiment. The dots in Fig. 2(a) represent a fit to an intermediate case of $\gamma \approx 1.8$. For both cases, ΔB is found to lie in the 13–15-G region. This residual linewidth includes homogeneous contributions from power broadening, pressure broadening, and natural linewidth. Other broadening mechanisms in our residual linewidth include magnetic-field inhomogeneity and instability, and Stark broadening due to bombarding electrons and space charge.

As an example of the possible precision of this method, we have determined from Fig. 2(b) the point corresponding to the center of the homogeneous transition. We find $B_0 = 71\,677 \pm 3$ G at a laser frequency of 29 511 068 MHz.[7] This corresponds to a determination, at this magnetic field, of the $9^1P(M_L = +1) \rightarrow 7^1S$ interval to a precision of about 0.1 ppm. Combination of a complete Zeeman-tuning theory and measurements now in progress at other fields should yield a comparable precision in the zero-field interval. We can further utilize this new technique to determine homogeneous linewidths, to resolve minute structure, and to measure many other atomic or molecular energy intervals to high precision.

We wish to acknowledge helpful discussions with R. Panock and the help provided by Larry Rubin in instrumentation.

The work done at the Francis Bitter National Magnet Laboratory was supported by the National Science Foundation. One of us (T.A.M.) is a Guest Scientist at the Francis Bitter National Magnet Laboratory.

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