Direct Optical Resolution of the Recoil Effect Using Saturated Absorption Spectroscopy

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The recoil-induced spectral doubling of the CH_4 saturated absorption peaks at 3.39 μm has been clearly resolved. A working resolving power of 8×10^{10} (1 kHz half width at half-maximum), achieved with aberration-compensated parabolic optics of 32 cm diam, was available to study the height ratio and splitting of the recoil peaks. The resolved Doppler-generated level crossings are not recoil doubled and give improved hfs information. We derive new, exact equations relating the atomic natural frequency to laboratory resonance frequencies.

As predicted by Kol'chenko, Rautian, and Sokolovskii,[1] momentum exchange between the radiation field and a quantum absorber gives rise to spectral doubling in saturation spectroscopy. The smallness of that frequency splitting may provide an ultimate natural challenge to the techniques of saturated-absorption spectroscopy. The CH_4 transition at 3.39 μm is a favorable case for this investigation in view of the small mass and high transition frequency. On the other hand, structure within the linewidth can only be regarded as detrimental to the otherwise very attractive prospects for an optical frequency standard. With suitable attention to the laser frequency stability and to the various spectral broadening mechanisms,[2] it has become possible to resolve[3] the two components clearly for the first time and to begin investigation of their pressure and power dependence.

We begin by recalling the physical origin of the recoil effect. We then mention the improvements of the laser spectrometer which led to this new level of resolution, and consider the several residual broadening mechanisms that limited the working resolution. Finally, we report improved frequency intervals for the CH_4 hyperfine spectrum.

An essential feature of sub-Doppler spectroscopy is the necessity to consider momentum conservation between the light and the absorbing molecules, as well as energy conservation: In fact the usual Doppler broadening is just a manifestation of momentum conservation. We also know that the resolution limit in sub-Doppler spectroscopy depends directly on how precisely the light momentum is defined.[2,4] A second manifestation of momentum conservation—not so familiar in optical spectroscopy—is the recoil effect. Although the recoil energy can be obtained from a nonrelativistic approach, a complete and consistent formulation including both the recoil and the relativistic Doppler effect can be developed by making a relativistic energy and momentum balance. In that way we can obtain the resonance condition for light of frequency ω and wave vector $\vec{k}$ applied to the two-level atom of transition frequency $\omega_0 = 2\pi\nu_0$. The atom's mass M may conveniently be referenced midway between the two atomic levels. For an absorber having a velocity $\vec{v}_a$ in its lower state, one derives the absorption resonance condition

$$\frac{\omega}{\omega_0} = \frac{(1 - v_a^2/c^2)^{1/2}}{1 - \vec{k}\cdot\vec{v}_a/\omega}\left(\frac{1}{1-\epsilon}\right),$$

where $\epsilon = \hbar\omega_0/2Mc^2$. For emission from the upper state, the resonance condition is

$$\frac{\omega}{\omega_0} = \frac{(1 - v_b^2/c^2)^{1/2}}{1 - \vec{k}\cdot\vec{v}_b/\omega}\left(\frac{1}{1+\epsilon}\right),$$

where $\vec{v}_b$ is the velocity in the upper state. These expressions correspond to single-quantum transitions. In saturation spectroscopy the molecules in a given velocity group must be able to interact with two waves of opposite wave vectors to suppress the Doppler broadening. For example, the velocity class corresponding to the population hole created in the lower state by absorption of the $+\vec{k}$ wave must also satisfy the absorption resonance condition for the probing $-\vec{k}$ wave.[2] This implies $\vec{k}\cdot\vec{v}_a = 0$ and the lower-state resonance condition

$$\frac{\omega}{\omega_0} = \left[1 - \left(\frac{v_a}{c}\right)^2\right]^{1/2}\frac{1}{1-\epsilon}.$$

The velocity-resolved population peak created in the excited state gives amplification of the second beam when the emission resonance condition is satisfied. This occurs when $\vec{k}\cdot\vec{v}_b = 0$ and

$$\frac{\omega}{\omega_0} = \left[1 - \left(\frac{v_b}{c}\right)^2\right]^{1/2}\frac{1}{1+\epsilon}.$$

To a good approximation we have $\omega/\omega_0 = 1 - v^2/2c^2 \pm \epsilon$. Thus one expects that in saturated absorption spectroscopy each spectral "line" will ordinarily be a doublet of splitting $2\delta = 2\epsilon\nu_0$, where the higher frequency component corresponds to saturation resonance in the lower state.[5] In the plane-wave approach of Kol'chenko, Rautian, and Sokolovskii,[1] the two peaks are Lorentzians of the same width and their heights are proportional to the respective reciprocal level decay rates. If the Gaussian spatial structure of the laser beam is taken into account, one obtains more complex amplitudes and shapes for the two peaks.[4] Some further theories describing the strong-field modifications of this picture have been presented and others are being developed.[6-10] If there is structure (e.g., hfs) within the Doppler width, one can observe three-level resonances (Doppler-generated level crossings) in saturated-absorption spectroscopy. (They arise from molecules with nonzero axial velocity.) Since only molecules in the one common level can interact with both light beams, we must expect only one recoil peak with a red or blue shift equal to δ, according to whether the common level is the upper or lower state.

Previous high-resolution experimental studies have provided through line-shape analysis some evidence for the recoil spectral doubling.[2] These experiments were limited to 2.5-kHz resolution by transit-time broadening, even with the absorption cell cooled to 77°K. The offset-locked spectrometer now has a new 13-m external absorption cell with astigmatically compensated,[11] internal, $f/10$ parabolic mirrors of 32-cm aperture. Wave-front aberrations are less than $\lambda/5$. The present 11-cm laser-beam radius should yield a transit broadening of $\simeq 650$ Hz half width at half-maximum (HWHM). Natural lifetime and residual Zeeman effect contribute less than 100 Hz each. The effective laser spectral width is about 200 Hz, but long-term drift of the reference system degraded the resolution another ~ 100 Hz over the 36-h integration period leading to the derivative data of Fig. 1 (lower curve). The normal (nonderivative) absorption spectrum formed from a 1-h sample of these data is shown (upper curve in Fig. 1) to help illustrate the incipient separation of the recoil doublets into pairs of distinct peaks. For the ~ 70-μTorr average pressure corresponding to Fig. 1, the collision-induced broadening is about 300 Hz. Data were taken for pressures between 10 and 300 μTorr; the base pressure was ~ 3 μTorr. For computer least-squares fitting of

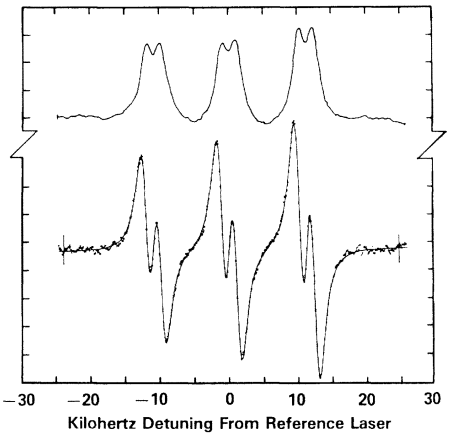

FIG. 1. Derivative spectrum of the three main hyperfine lines of $^{12}C_4$ showing the recoil doubling (lower curve). Methane pressure, 70 μTorr; room temperature; modulation, 800 Hz peak to peak. A least-squares fit (solid line) gives a width of 1.27 kHz HWHM and a recoil doublet splitting of 2.150 kHz, the the high-frequency peak 1% larger than the low-frequency ones. The upper curve, integrated from a sample of such data, shows that each hyperfine component is spectrally doubled by the recoil effect.

such spectra we assumed a line shape consisting of three pairs of modulation-broadened Lorentz functions,[12] which is a reasonable approximation when the transit-time broadening and finite-lifetime broadening are comparable.[4] For all three hyperfine components we imposed common widths, and common recoil doublet height ratios and splittings. This least-squares fit, as represented by the solid line in Fig. 1, gives a linewidth (unmodulated) of 1.27 kHz HWHM. The recoil doublet splitting is 2.150 ± 0.005 kHz compared with the expected theoretical value 2.163 kHz (see below). The height ratio is (0.989 ± 0.003):1. Half-widths as low as 990 Hz were observed for pressures in the 10-μTorr range, with a broadening rate of 4.2 ± 0.5 Hz/μTorr. At these low pressures, the height ratio decreased by a few percent. This less than unit ratio is understandable in view of the finite radiative lifetime of the excited state and the enhanced contribution of slow molecules.[4] At higher pressure ($\geq$ 100 μTorr) the height ratio is fixed by the ratio of the two collision decay constants which describe the loss of velocity-resolved information in each state. Experimentally we found a height ratio within 2% of 1.00:1.

At higher intensities, the observed doublet splitting has a tendency to be smaller (~ 1%). This effect can be understood as a three-photon process and was studied with a fifth-order expansion

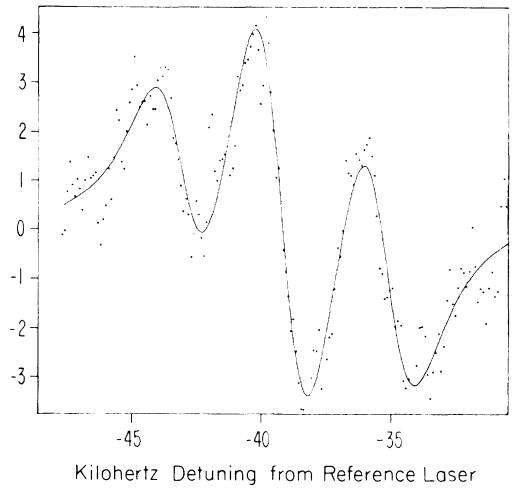

FIG. 2. Derivative spectrum of the four Doppler-generated level crossings between the $\Delta F = \Delta J = -1$ and $\Delta F = 0$ transitions. Central pair is unresolved. Fit function (solid line) is four recoil-shifted single Lorentzians with 1.7 kHz common width. Modulation, 1.3 kHz peak to peak; methane pressure, 100 μTorr.

in both fields,[8] and with a calculation which treats one field to all orders and the probe field to first order.[9]

In Fig. 2 we show the spectral region of the Doppler-generated level crossings between the strong $\Delta F = \Delta J = -1$ and the weaker $\Delta F = 0$ transitions.[13] We note that these transitions are not recoil-doubled and consequently may be of interest for optical frequency standards of improved accuracy using suitable molecules. By use of the data of Figs. 1 and 2 it is possible to improve our knowledge of the methane hyperfine energy levels. Using the notation of Ref. 13 and a number of spectra similar to Fig. 1, we find the main peak positions to be $A - a = +11.06$ kHz for the high-frequency $(v_3, J, F) = (0, 7, 8) \rightarrow (1, 6, 7)$ transition. For the low-frequency $(0, 7, 6) \rightarrow (1, 6, 5)$ transition we have $b - B = -11.34$ kHz. Both spectral positions have 50-Hz uncertainties. The $(0, 7, 7) \rightarrow (1, 6, 6)$ line is referenced to zero. For the crossing resonances we obtain $-A/2 - \delta = -35.3$ kHz, $-A + a/2 + \delta = -38.7$ kHz, $-b + B/2 - \delta = -39.7$ kHz, and $-b/2 + \delta = -43.2$ kHz, all with 500-Hz uncertainties. The inferred ground-state and excited-state hyperfine intervals are now within 200 Hz and 5 kHz, respectively, of the theoretical values.[14]

In conclusion, this paper presents direct evidence for the existence of the recoil spectral doublet structure in saturation spectroscopy and therefore provides a first quantitative demonstration of the quantized momentum exchange between a free-flying molecule and the light field. For the methane transition studied we find height ratios close to 1:1 with a small enough pressure dependence that frequency standards based on heavy molecules such as OsO_4 may be expected to give good results even without complete resolution of the recoil structure.

It is also interesting to note that the recoil splitting provides a measure of h/M in frequency units. With uv transitions in light molecules or even with the Ca 6573-Å transition,[15] it may be possible to make an interesting measurement of this quantity.

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The following Letter should have appeared in the 1 November 1976 issue. We regret that a misunderstanding resulted in publication of reference material instead of the submitted manuscript. See Erratum, this issue, page 1368.

Strong-Signal Theory of a Free-Electron Laser*

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The strong-signal regime of a free-electron laser is analyzed in terms of a set of "generalized Bloch equations." We show that for current free-electron-laser configurations the saturation will be reached for a field on the order of 10^7 V/m, with an efficiency at saturation of 5×10^{-3}. However, a strong reshaping of the electron distribution may alter the efficiency of free-electron lasers in cases where the electron beam is recycled from one shot to the next.

Recently, a considerable effort has been made toward the realization of a free-electron laser. Elias *et al.*[1] have passed a relativistic electron beam ($E=\gamma mc^2\simeq 24$ MeV) through a helical static magnetic field, and have observed stimulated scattering at 10.6 μm. On the theoretical side, a number of authors[2-5] have computed the small-signal gain of this laser, and there is now general agreement on the functional form as well as the numerical value of the gain. In order to assess the potential of any practical laser device, it is necessary to *complement* the small-signal theory by an analysis of the mechanisms of saturation. In the present case, we do not need a quantum theory.[5] In fact, the quantum theory of a free-electron laser is extremely tedious, and neither *desirable* nor necessary.

In this Letter, we present the strong-signal theory of a free-electron laser. Our analysis is completely classical and relies on the coupling of Maxwell's equations to the relativistic collisionless Boltzmann equation.[5] We use the Weizsacker-Williams approximation,[6] which allows us to simulate the static magnetic field of period λ_q by a fictious incident electromagnetic field of wavelength $\lambda_i=(1+v/c)\lambda_q\simeq 2\lambda_q$, propagating in the opposite direction of the electron beam. We then express the problem in terms of a set of generalized Bloch equations coupled to Maxwell's equations.

Following the derivation of Ref. 5, we find that the coupled Maxwell-Boltzmann equations can be reduced to the set of equations

$$\left(\frac{\partial^2}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{A}_T=\frac{e^2\mathfrak{F}}{mc\epsilon_0}\vec{A}_T\int_{-\infty}^{\infty}dp_z\frac{h(z,p_z,t)}{\gamma}, \quad (1)$$

$$\frac{\partial h}{\partial t}+\frac{p_z}{m\gamma}\frac{\partial h}{\partial z}=\frac{e^2}{m\gamma}\frac{\partial}{\partial z}\left(\frac{\vec{A}_T^{\,2}}{2}\right)\frac{\partial h}{\partial p_z}. \quad (2)$$

Here, the electrons have been taken to be injected inside the cavity along the z axis. $p_z=\gamma mv_z$ is the z component of the electron momentum, and $\gamma\simeq[1+(p_z/mc)^2]^{1/2}$. The filling factor $\mathfrak{F}$ is the ratio of the section πa^2 of the electron beam to the section of the cavity. $h(z,p_z,t)$ is the longitudinal part of the Boltzmann distribution function and is related to the electron number $N(t)$ inside the cavity by

$$N(t)=\pi a^2\int_{-\infty}^{\infty}dp_z\int_0^L dz\,h(z,p_z,t), \quad (3)$$

where L is the length of the cavity.

$\vec{A}_T$ is the vector potential that we take to be of