PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Laser Heterodyne Spectroscopy [and Discussion]

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Laser heterodyne spectroscopy

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The principles and application of heterodyne techniques to optical and infrared spectroscopy are discussed. Examples are drawn from the field of atomic isotope shifts and hyperfine structure and from molecular spectroscopy. Measurement of beat frequencies between two laser beams is considered, but also the detection of incoherent radiation by employing a laser local oscillator is reviewed. Applications in astronomy provide examples of the latter technique.

Introduction and basic principles

Heterodyne techniques have been in general use in radio engineering for many decades. In a typical radio receiver a weak incoming signal is mixed in a nonlinear circuit element with a relatively strong local oscillator signal. The difference frequency, or intermediate frequency, is selected by filters and amplified in a narrow-band fixed-frequency amplifier. There are many advantages of the technique, not the least being that it is convenient to control the gain and selectivity of the fixed-frequency stages. Furthermore, intermediate frequencies can be chosen to fall in regions of the spectrum where useful amplifying devices are readily available. This feature is of great importance in frequency measurement. Direct electronic counters function up to about 500 MHz only, but highly accurate microwave-frequency meters are available in which the unknown signal is mixed with a known harmonic of a stable frequency source such that the heterodyne beat falls within the range of counters.

It was Forrester (1961) who first suggested that a laser would be a suitable local oscillator for an optical heterodyne system, following upon his demonstration of heterodyne beats between incoherent sources. Given a suitable local oscillator and mixer it should be possible to transfer the same advantages into the optical spectrum as are familiar in the radiofrequency régime. Most optical detectors are square-law devices, because the response is proportional to the intensity of illumination. They are thus ideal heterodyne beat detectors, assuming that the two beams of light may be appropriately mixed and that the detector has an adequate bandwidth to respond to the beat frequency. Suppose that two perfectly aligned plane waves of amplitudes A_1 and A_2 and angular frequencies ω_1 and ω_2 are incident upon a detector, then the individual disturbances may be represented by fields E_1 and E_2 such that

$$E_1 = A_1 \cos \omega_1 t$$

and

$$E_2 = A_2 \cos (\omega_2 t + \phi).$$

Individually these waves would yield measured intensities

$$I_1 = A_1^2$$

and

$$I_2 = A_2^2.$$

However, the resultant intensity when the two waves are detected simultaneously is

$$I_3 = I_1 + I_2 + 2\sqrt{(I_1 I_2)} \cos \{(\omega_2 - \omega_1)t + \phi\},$$

where we have assumed that the detector has an ideal square-law response. If indeed the detector is able to respond to the frequency $(\omega_2 - \omega_1)$ then the amplitude of the beat signal seen in the response of the detector is just $2\sqrt{(I_1 I_2)}$. It is also worthy of note that any phase inform a-tion, embodied in ϕ, propagates through to the beat signal.

The simple treatment above assumed perfect alignment of two light beams. If two such plane waves are not perfectly aligned, but a very small angle θ exists between the wave fronts, then the heterodyne signal will be reduced. A small detector will see interference fringes propagating across its surface at the beat frequency. Clearly if there is to be a reasonable heterodyne signal, the detector linear size, d, must be significantly less than the fringe width. This means that the alignment must be such as to ensure that

$$\theta < \lambda/d,$$

where λ is the wavelength of the radiation. Alternatively if two beams with Gaussian intensity distributions are mixed on a detector of relatively large area the same relation must be satisfied, with d taking on the significance of the beam size. A useful review of these general considerations and other aspects of optical heterodyne receivers has been written by DeLange (1968).

If the beat frequency is to be observed between two laser beams emerging from similar devices then it is not difficult to satisfy the alignment criterion above. Such methods are now widely used in atomic and molecular spectroscopy, and a number of examples will be given in the next section. On the other hand, if a laser is to be used as a local oscillator to detect in-coherent radiation from a thermal source a rather different situation exists, which is of con-siderable interest in astronomy. A number of authors have pointed out that the optical hetero-dyne detector acts simultaneously as an antenna and as a mixer, but there is some confusion in the literature as to the sensitivity of such a device for astronomical purposes. Some examples will be given in the third section of this paper.

Applications of the heterodyne technique in atomic and molecular spectroscopy

The development of tunable continuous-wave lasers and of several techniques of sub-Doppler spectroscopy caused a remarkable awakening of interest in atomic and molecular spectroscopy in both visible and infrared spectral regions. For the first time it became possible to scan spectral features with high instrumental resolution and with excellent signal strength. In the realm of atomic spectroscopy, for example, hyperfine splitting and isotope effects could be easily resolved, down to the natural line width. Many sub-Doppler techniques have been devised, but one particularly simple method involves producing a well collimated beam of atoms and causing the beam from a tunable laser to intersect this at right angles. There is then no first-order Doppler shift of the interaction between light and atoms. If scattered light is observed from the interaction region and the laser is scanned through an atomic resonance, essentially Doppler-free spectra will be observed. Such methods have been widely used for the study of optical isotope shifts (Broadhurst 1974; Griffith 1979, 1981; New 1981; Baird 1979; Bekk 1979) and hyperfine structure (see, for example, Griffith 1977; Brand 1981). A typical spectrum is shown in figure 1.

In general, experiments of this kind seek to measure the separation of components in the spectrum, so that some kind of frequency calibration of the laser scan is necessary. A simple

and convenient method is to pass a fraction of the laser beam through a confocal Fabry–Perot interferometer, which will produce frequency markers separated by its free spectral range. However, it is only with great difficulty that absolute accuracies much better than ± 1 MHz may be obtained this way, on account of the drifts and nonlinearities of lasers and etalons. Bearing in mind that spectral line widths may be only a few megahertz and that signal:noise ratios are usually excellent it would be reasonable to expect far superior accuracy. Heterodyne methods belong to one class of solution to this problem.

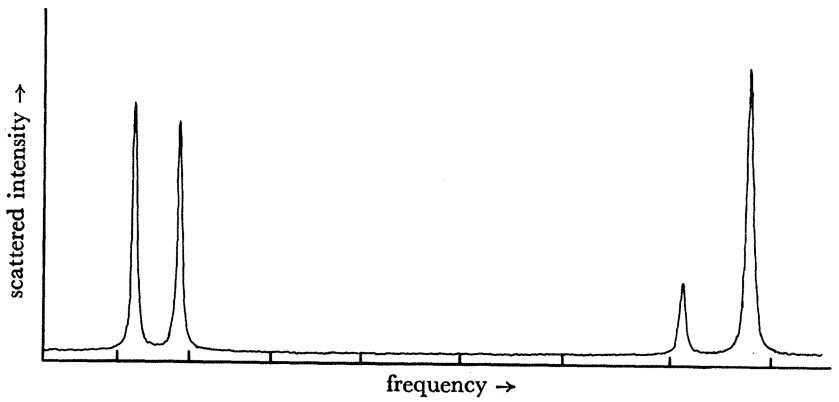

FIGURE 1. A typical spectrum yielded by laser spectroscopy. It was obtained by scanning a single laser through the hyperfine structure of the 589.6 nm transition of sodium in an atomic beam. The ticks on the abscissa are 300 MHz apart and show the positions of marker points derived by passing a fraction of the laser beam through a Fabry–Perot etalon.

A second practical difficulty with laser spectroscopy may appear when a search is being conducted for a very weak spectral feature. A suitable approach would consist of scanning the spectral region of interest repeatedly, integrating data over a long time; however, this requires a defined frequency space to be scanned reproducibly. Natural drifts of lasers and interferometers can make this very difficult unless a heterodyne approach is employed to establish a frequency scale.

There are several ways of employing the heterodyne principle in laser spectroscopy. The general technique adopted at Birmingham is illustrated in figure 2. Two continuous-wave tunable dye lasers were arranged so that their beams could be mixed collinearly in a beam splitter. This mixing was achieved with an accuracy of alignment consistent with the criterion given above. One of the mixed beams intersected the atomic beam at right angles, whereas the second was focused on to the sensitive area of a wideband silicon avalanche photodiode. Beat signals detected by this photodiode were amplified by using broadband amplifiers so that the signal was strong enough to drive a microwave frequency meter directly. This frequency meter was interfaced to a computer, which could thus acquire a series of measurements of the heterodyne beat frequency.

In operation each laser was locked by means of a servo loop to the line centre of a component of the spectrum being studied. The servolocking technique consisted of deliberately modulating the laser frequency and detecting the modulation frequency as intensity modulation of a scattered light signal. When the laser was centred on a spectral peak there was no fundamental component of the modulation rate in this signal. Phase-sensitive rectification served to yield an

appropriate error signal. The two lasers were modulated at different rates so that independent locking of each one to separate features was possible. When so locked, the detected heterodyne beat frequency gave the separation of the two features directly. In practice numerous frequency measurements were made and statistically averaged. Details have been reported by Griffith (1977).

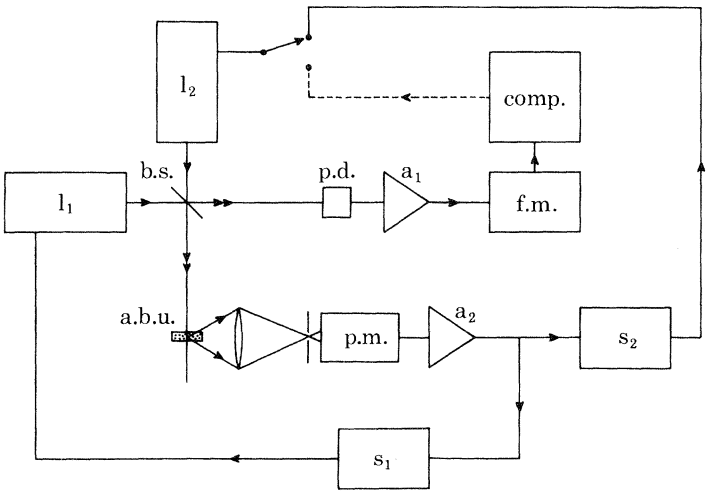

FIGURE 2. A schematic diagram of one version of the Birmingham laser heterodyne system. The beams from two tunable dye lasers, l_1 and l_2, are combined in the beam splitter, b.s. One combined beam passes to the wideband photodiode, p.d., which yields a heterodyne beat signal. This signal is amplified by a_1 and the frequency is measured by the meter, f.m., which passes the result to the computer, comp. The second combined beam goes to the atomic beam unit, a.b.u. Scattered light is detected by the photomultiplier, p.m., the current from which is amplified by a_2. Servo loops s_1 and s_2 may be used to lock the laser frequencies to atomic transitions. Alternatively the link shown dotted may be used to establish a heterodyne offset servo loop.

This system was used to study the hyperfine splitting of sodium (Griffith 1977) and the optical isotope effect of samarium (Griffith 1979; New 1981; Griffith 1981). For sodium, uncertainties of ± 20 kHz were obtained on the splitting of the $3P_{\frac{1}{2}}$ level (188.93 MHz). The splitting of the ground state ($3S_{\frac{1}{2}}$) was also measured as a test of the technique, since this parameter was already known with an uncertainty of about ± 1 Hz from atomic beam r.f. spectroscopy. This latter splitting, which is about 1.8 GHz, was found to agree with expectations within our uncertainty of 20 kHz. Measurements of samarium isotope shifts of the order of 1 GHz were made with uncertainties of 200 kHz, which revealed the existence of anomalies in these shifts.

The experiments mentioned above were made with unstabilized dye lasers showing substantial spurious frequency modulation due to acoustically driven vibrations of the laser mirrors. Typical laser line widths of greater than 10 MHz were seen. Work is currently in hand on frequency-stabilized dye lasers in which another electronic servo loop is used to reduce the line width. It is expected that this will yield a substantial improvement in the precision of the frequency measurements. The method described above, in which each laser is held continuously on resonance with an atomic transition, has the benefit of making the most effective use of experimental time, but it has the disadvantage that it does not allow line profiles to be scanned. Exploring the profile is clearly essential when spectral lines have overlapping components.

A solution to the problem of scanning a laser reproducibly is offered by the technique called frequency offset locking. On the schematic diagram of figure 2, an alternative frequency-

locking servo loop is shown by a dotted line. The principle of using this is as follows. One laser was locked, as before, to an atomic spectral line either in the common atomic beam, or a separate one. The second laser was then set to have a defined heterodyne difference frequency separating it from the first. This was achieved by completing a servo loop including the computer, which sent signals to the second laser as required to achieve the desired beat frequency. Since this beat frequency could be set by the computer it was possible to scan the second laser reproducibly through a range of difference frequencies. By this technique it is possible to explore complicated spectral structure, or to search consistently in one region of frequency space seeking a weak signal.

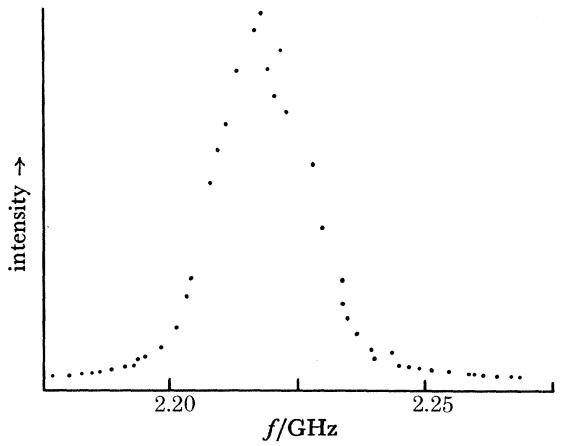

FIGURE 3. The peak shows the resonant scattering intensity from ^{144}Sm present in an atomic beam of Nd at the level of 6 atoms per million. A reference laser was locked to the corresponding peak of ^{148}Sm in a separate Sm beam. A second laser was offset in frequency from the reference by controlled amounts chosen to scan only the region where ^{144}Sm was expected to lie.

An example of such a case is given in figure 3 which shows a resonance peak of the isotope ^{144}Sm present at the level of only 6 atoms per million in an atomic beam of Nd. A similar technique has been employed by a group from Karlsruhe in their work on unstable barium isotopes (Nowicki 1978; Bekk 1979). Again the ability to produce well calibrated, reproducible scans was important to their work, where they were operating with minute radioactive samples. They were able to make measurements of isotope effects with uncertainties of less than 1 MHz starting with samples as small as 40 pg for nuclei of as little as 11 min half-life by using otherwise conventional atomic beam techniques.

Heterodyne methods similar to those described above are now finding widespread use in the infrared region, where the interest is in the study of molecular rotation–vibration spectra. CO_2 lasers and tunable diode lasers particularly have found applications in this area: for example, in one experiment (Wells 1979) a series of infrared calibration frequencies was established by using the spectrum of carbonyl sulphide. A CO_2 laser was stabilized to a fixed frequency by locking it to a saturated absorption dip. A tunable diode laser was locked to successive peaks in the spectrum of OCS and the beat frequency was measured in each case between the two laser beams. A microwave spectrum analyser was used for this measurement. The object of the experiment was to produce well defined frequency markers for tunable diode laser users.

A most important application of heterodyne techniques in the infrared and visible regions has been in establishing precise optical frequency standards. In a series of remarkable

experiments in standards laboratories, notably the National Physical Laboratory, Teddington, N.R.C., Canada, and the National Bureau of Standards, Colorado, it has been demonstrated that the frequencies of carefully stabilized lasers may be referred back to the fundamental time standard. This was achieved by multiple heterodyne stages with numerous laser types. No more will be said on this as a separate paper is being contributed to these proceedings on the topic; however, a summary has been written by Evenson (1977).

Recently a new form of heterodyne spectroscopy has been reported (Hall 1981; Bjorklund 1981) in which a single laser may be used. The general principle comprises the use of phase modulation of a single-frequency laser beam by an electro-optical modulator. Conditions are arranged so that the rate of modulation is large, but the modulation index is small. The modulated frequency spectrum then consists of a strong unshifted line and two weak sidebands symmetrically placed on either side, separated from the main peak by the modulation frequency. If this beam is allowed to fall on a high-speed detector there will be no observed beating between the frequencies because the amplitude and phase of the sidebands is such as to cancel heterodyne beats exactly. This is self-evident because pure phase modulation produces no amplitude modulation.

If a laser beam, modulated as described above, is passed through, say, a gaseous absorption cell and tuned so that one sideband is nearly on resonance but the other is not, then the situation is changed. The phase and amplitude of the one sideband will be modified relative to the other by virtue of its passage through a nearly resonant medium. As a result a heterodyne beat will be observed in the detector. This signal may be demodulated in a phase sensitive rectifier operating at the modulation frequency. Analysis shows that two signals may be derived, a normal absorption signal if an in-phase reference signal is used and a dispersive absorption curve if a quadrature reference is employed. Several advantages have been claimed for the techniques. Hall points out that the modulation frequencies used are so high (of the order of 10 MHz) that they lie well above the frequencies of amplitude noise generated by dye lasers. Detection sensitivities limited only by shot noise are therefore possible. Within the limitations of existing modulators it is also possible to vary the effective laser frequency by changing the modulation frequency rather than the laser centre frequency.

In all of the examples discussed so far there is no great practical difficulty introduced by the problems of alignment or of signal strength. Working directly with laser beams ensures a well collimated source of almost arbitrary brightness. There are certain limitations in the performance of detectors, however. Commercial p-i-n avalanche photodiodes may be effectively used up to about 5 GHz beat frequencies. Sensitive areas are usually very small (about 0.05 mm^2), but laser beams are easily focused to suitable spot sizes. Mixing of laser beams on metallic point contact 'cat's whisker' diodes has been used by the standards laboratories to achieve immensely greater bandwidths. Nonlinear optical media may also be used for frequency conversion.

Heterodyne detection of incoherent and thermal sources

An optical receiver for incoherent radiation such as might come from astronomical objects would seem to be an attractive proposition. Very high spectral resolution and very stable frequencies can be achieved. However, there are plainly problems in mixing the incoming radiation with that from the laser local oscillator. The alignment criterion must still be met so that the effective angular acceptance of the antenna of such an optical receiver is very small.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

For stellar objects this would present no problem and the heterodyne method apparently offers a very efficient way of detecting a weak source. However, considerable discouragement was provided in a well known paper by Siegman (1966), where the antenna properties of an optical heterodyne receiver were discussed and this was extended to the consideration of the signal: noise ratio to be expected when detecting thermal radiation.

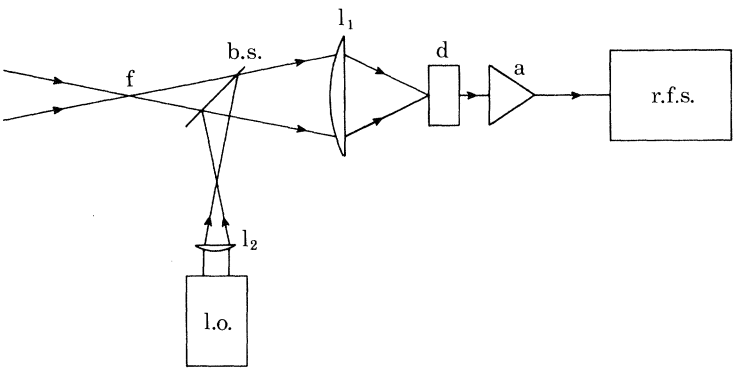

FIGURE 4. This is a simplified view of the astronomical infra red heterodyne receiver used by Betz. Light from a telescope is brought to a focus (f) and then passes through a beam splitter (b.s.), where it is mixed with radiation from the local oscillator laser (l.o.). The lenses l_1 and l_2 were used to match the two beams geometrically and to focus them on the detector (d). Heterodyne signals from the detector were amplified (a) and passed to a radiofrequency spectrometer r.f.s.

Siegman's conclusion was that heterodyne receivers might be useful in the infrared, but not in the visible spectrum. Indeed he stated that a visible light receiver pointed directly at the Sun would show no detectable signal above noise. However, this conclusion has been shown to be incorrect because it ignored the possibility of integrating data-taking over extended periods of time. This was graphically demonstrated by Veth (1973) who detected solar radiation at 633 nm with a heterodyne receiver, achieving good signal:noise in line widths of the order of 0.1 pm. Similar detection of visible radiation from bright stars has also been reported (Nieuwenhuijzen 1970). Nevertheless little use has been made of the method in the visible part of the spectrum: most progress has been seen in the infrared region.

Several early experiments to demonstrate the feasibility of heterodyne detection of thermal radiation used CO_2 lasers providing local oscillator radiation at 30 THz. Detection of black body radiation in the temperature range 300–900 K was reported by Gay (1973). Several relevant papers were presented to the Workshop on Coherent Detection in Optical and Infrared Astronomy (van Bueren 1975). This included a useful review of signal:noise considerations (Blaney 1975). One of the earliest applications of the method in astronomy was a study of the carbon dioxide on Mars (Betz 1975, 1977), which showed well resolved absorption lines. Emission lines from Venus were reported in the same work.

The work of Betz has been carried out with a heterodyne receiver installed on a telescope at Kitt Peak National Observatory. A simplified diagram illustrating the equipment is shown in figure 4. The beam from a CO_2 or N_2O laser is shaped by a lens to match the geometrical properties of the telescope beam close to its main focus. Combination of the two beams takes place at a beam splitter and another lens produces a common focus on a HgCdTe photodiode. This is followed by an amplifier with a bandwidth of 3.2 GHz. After further processing the beat signals are analysed in a parallel bank of 128 filters giving adjoining channels 20 MHz wide, a

figure representing the instrumental resolution of about 1 in 10^6. The overall detection performance is reported to be limited only by quantum noise (Betz 1981). By using this spectrometer the existence of ammonia and ethylene in the gas surrounding supergiant stars has been unambiguously demonstrated. In general the heterodyne technique appears to offer the best signal:noise ratio for detection of 10 μm radiation when a resolution of better than about 1 in 10^5 is needed. A further bonus lies in the precise calibration of the absolute frequency scale and the reproducibility of this scale from time to time. The scale can be arranged to track Doppler shifts if required.

Summary

Optical heterodyne spectroscopy has been shown to permit measurements with high resolution, low noise and unsurpassed accuracy in several fields. There seems little doubt that variants on the technique will become of increasing importance. It is perhaps especially true that its impact will be felt as the ability to locate the centres of spectral features improves. Recent work on cooled trapped ions (Dehmelt 1981) suggests that it is possible to observe extremely narrow spectral lines corresponding to long-lived states and that, perhaps, spectral resolution of 1 part in 10^{18} may one day be achieved. Clearly such figures imply the need for extraordinary stability and control of laser sources. At present it would seem that the direct frequency counting which is a feature of heterodyne systems offers the only possibility of approaching the required quality.

In astronomy one looks to the extension of heterodyne methods outside the 10 μm band. Of possibly great interest is the preservation of phase in the heterodyne signal, mentioned earlier. This offers the prospect of using interferometric techniques by employing two or more detectors used coherently, as is now commonplace in radio astronomy.

I gratefully acknowledge the work of R. New, G. R. Isaak, M. P. Ralls and G. M. Hayes on the heterodyne experiments performed at Birmingham as described above, and the technical expertise of J. W. Litherland, which was invaluable in many of the experiments mentioned.

References

Baird, P. E. G., Brambley, R. J., Burnett, K., Stacey, D. N., Warrington, D. M. & Woodgate, G. K. 1979 *Proc. R. Soc. Lond.* A **365**, 567–582.

Bekk, K., Andl, A., Goring, S., Hanser, A., Nowicki, G., Rebel, H. & Schatz, G. 1979 *Z. Phys.* A **291**, 219–230.

Betz, A. L. 1975 *Space Sci. Rev.* **17**, 659.

Betz, A. L. 1981 In *Laser spectroscopy V* (ed. A. R. W. McKellar, T. Oka & B. P. Stoicheff), pp. 81–88. Berlin: Springer-Verlag.

Betz, A. L., Sutton, E. C. & McLaren, R. A. 1977 In *Laser spectroscopy III* (ed. J. L. Hall & J. L. Carlsten), pp. 31–38. Berlin: Springer-Verlag.

Bjorklund, G. C., Lenth, W., Levenson, M. D. & Ortiz, C. 1981 In *Laser spectroscopy V* (ed. A. R. W. McKellar, T. Oka & B. P. Stoicheff), pp. 389–398. Berlin: Springer-Verlag.

Blaney, T. G. 1975 *Space Sci. Rev.* **17**, 691–702.

Brand, H., Pfeufer, V. & Steudel, A. 1981 *Z. Phys.* A **302**, 291–298.

Broadhurst, J. H., Cage, M. E., Clark, D. L., Greenlees, G. W., Griffith, J. A. R. & Isaak, G. R. 1974 *J. Phys.* B **7**, L513–L517.

Dehmelt, H. 1981 In *Laser spectroscopy V* (ed. A. R. W. McKellar, T. Oka & B. P. Stoicheff), pp. 353–359. Berlin: Springer-Verlag.

DeLange, O. E. 1968 *I.E.E.E. Spectrum*, October, pp. 77–85.

Evenson, K. M., Jennings, D. A., Petersen, F. R. & Wells, J. S. 1977 In *Laser spectroscopy III* (ed. J. L. Hall & J. L. Carlsten), pp. 56–68. Berlin: Springer-Verlag.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Forrester, A. T. 1961 *J. opt. Soc. Am.* **51**, 253–259.
Gay, J., Journet, A., Christophe, B. & Robert, M. 1973 *Appl. Phys. Lett.* **22**, 448–449.
Griffith, J. A. R., Isaak, G. R., New, R. & Ralls, M. P. 1981 *J. Phys.* B **14**, 2769–2780.
Griffith, J. A. R., Isaak, G. R., New, R., Ralls, M. P. & van Zyl, C. P. 1977 *J. Phys.* B **10**, L91–L95.
Griffith, J. A. R., Isaak, G. R., New, R., Ralls, M. P. & van Zyl, C. P. 1979 *J. Phys.* B **12**, L1–L7.
Hall, J. L., Baer, T., Hollberg, L. & Robinson, H. G. 1981 In *Laser spectroscopy V* (ed. A. R. W. McKellar, T. Oka & B. P. Stoicheff), pp. 15–24. Berlin: Springer-Verlag.
New, R., Griffith, J. A. R., Isaak, G. R. & Ralls, M. P. 1981 *J. Phys.* B **14**, L135–L139.
Nieuwenhuijzen, H. 1970 *Mon. Not. R. astr. Soc.* **150**, 325–335.
Nowicki, G., Bekk, K., Goring, S., Hanser, A., Rebel, H. & Schatz, G. 1978 *Phys. Rev.* C **18**, 2369–2379.
Siegman, A. E. 1966 *Proc. I.E.E.E.* **54**, 1350–1356 (also published in *Appl. Opt.* **5**, 1588–1594).
van Bueren, H. G. 1975 *Space Sci. Rev.* **17**, 621–628.
Veth, C., de Graauw, Th., Shelton, J. C. & van de Stadt, H. 1973 *Astron. Astrophys.* **26**, 479–482.
Wells, J. S., Petersen, F. R. & Maki, A. G. 1979 *Appl. Opt.* **18**, 3567–3573.

Discussion

G. Duxbury (*Department of Natural Philosophy, University of Strathclyde, Glasgow, U.K.*). The experiments described that make use of an electro-optic modulator to generate sidebands can also be carried out with an acousto-optic modulator. An a.o.m. can be designed to produce a large fraction of the pump laser power in the sideband, making it feasible to carry out saturation spectroscopy experiments by using the sideband itself. The spacing between the single sideband and the carrier can be up to *ca.* 500 MHz. Amplitude modulation of the sideband can be used to produce two frequencies symmetrically related to the position of the unsplit sideband. If the main laser is locked, and the modulator frequency is swept by using a computer-controlled frequency synthesizer, it is possible to get complete electronic tuning of the absolute frequency of the sideband. This method requires only a single laser, and reduces the laser stabilization problem to that of stabilizing a single device.

J. A. R. Griffith. Dr Duxbury has correctly drawn attention to a powerful technique for producing a controlled shift in a laser frequency. There are, however, some important physical differences between the type of experiment described and the work of Hall, for example, to which I referred in my talk. Hall uses the heterodyne signal, which only appears when one sideband is close to a resonance, as a sensitive signature for the detection of a resonant spectral feature. Interestingly the full experimental arrangement employed by Hall uses both the phase-modulation technique referred to in my talk and the acousto-optical shifting method described by Dr Duxbury. The saving of one laser is a valuable point provided that limited frequency coverage can be tolerated. Many experiments, including our own, require frequency ranges much greater than 500 MHz. By using two lasers and commercially available detection equipment a range of about 8 GHz is fairly easily obtainable.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES