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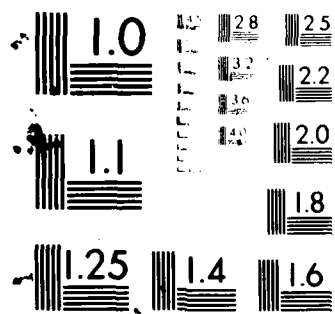
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MULTIPLE MODULATION FOR OPTICAL PUMP-PROBE SPECTROSCOPY.(U)
DEC 81 P BADO, S B WILSON, K R WILSON N00014-78-C-0325
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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1 REPORT NUMBER 5	2 GOVT ACCESSION NO. AD-A109591	3 RECIPIENT'S CATALOG NUMBER
4 TITLE (and Subtitle) MULTIPLE MODULATION FOR OPTICAL PUMP-PROBE SPECTROSCOPY		5 TYPE OF REPORT & PERIOD COVERED Technical
		6 PERFORMING ORG. REPORT NUMBER
7 AUTHOR(s) Philippe Bado, Scott B. Wilson and Kent R. Wilson		8 CONTRACT OR GRANT NUMBER(s) ONR-N00014-78 C-0325
9 PERFORMING ORGANIZATION NAME AND ADDRESS Department of Chemistry University of California, San Diego La Jolla, CA 92093		10 PROGRAM ELEMENT PROJECT, TASK AREA & WORK UNIT NUMBERS
11 CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research Arlington, VA 22217		12 REPORT DATE December, 1981
		13 NUMBER OF PAGES 5
14 MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15 SECURITY CLASS (of this report) Unclassified
		15a. DECLASSIFICATION DOWNGRADING SCHEDULE
16 DISTRIBUTION STATEMENT (of this Report) This document has been approved for public release and sale; its distribution is unlimited.		
17 DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report) DTIC SELECTED JAN 13 1982 E		
18 SUPPLEMENTARY NOTES Submitted to the Review of Scientific Instruments for publication		
19 KEY WORDS (Continue on reverse side if necessary and identify by block number) laser picosecond modulation		
20 ABSTRACT (Continue on reverse side if necessary and identify by block number) A system built largely from readily available amateur radio gear which uses radio frequency and audio modulation of pump and probe light beams allows the detection of very weak signals (rms noise $\sim 3 \times 10^{-11}$ W for a 10-second time constant) even in the presence of a much larger (3.6×10^{-2} W) background of pump and probe light.		

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TECHNICAL REPORT NO. 5

MULTIPLE MODULATION FOR OPTICAL PUMP-PROBE
SPECTROSCOPY

BY

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ABSTRACT

A system built largely from readily available amateur radio gear which uses radio frequency and audio modulation of pump and probe light beams allows the detection of very weak signals (rms noise $\sim 3 \times 10^{-11}$ W for a 10 second time constant) even in the presence of a much larger (3.6×10^{-2} W) background of pump and probe light.

Submitted to Rev. Sci. Instrum

PACS numbers 42.60.Fc, 78.40.-g, 78.30.-j

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MULTIPLE MODULATION FOR OPTICAL PUMP-PROBE SPECTROSCOPY

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A common type of spectroscopy involves pumping a sample with one light beam and probing the effect on the sample with a second beam, observing, for example, with a probe beam the change in absorption or scattering induced by a pump beam. An ideal experimental apparatus would discriminate against effects due to either the pump or probe beams acting alone. We point out in this note how this can be accomplished in a relatively simple manner using multiple modulation of the light beams. We demonstrate the technique with an example from picosecond transient electronic absorption using audio frequency (AF) amplitude-modulated (AM) radio-frequency (RF) modulation of the pump beam and RF modulation of the probe beam with observation at the difference frequency of the two RF signals, followed by synchronous AM demodulation. Performance near the noise limit which would be expected from a random photon arrival distribution (shot-noise limit) is achieved with relatively inexpensive and easily obtainable amateur radio equipment.

The instrumentation is shown schematically in Fig. 1. The pump and probe light beams are RF modulated by commercial electro-optical modulators¹ (Coherent Model 28) followed by polarizers as suggested for single RF modulation by Heritage, Levine and Bethea^{2,3} who have demonstrated that the noise in synchronously-pumped dye lasers falls off dramatically on going from the AF to the RF region, approaching the shot noise limit. The RF voltages are supplied by commercial amateur radio transmitters (Drake Model TR-7) impedance matched to the modulators through variable antenna couplers (Cubic Model ST-3B). The transmit inhibit line is cut on the TR-7 parent boards so they can operate anywhere from 1.5-30MHz so that the sum or difference frequency can be tuned to a minimum region in the laser noise spectrum. It is recommended that the modulators be well shielded inside copper boxes to insure against spurious radiation. While the present detection system is relatively immune to such radiation, one's neighbor's may not be. A 0-270 VDC bias is applied in parallel with the RF voltage, the DC voltage being blocked from the RF source by two 0.01 μ F capacitors and the RF voltage blocked from the DC supply with two 5 mH RF chokes.⁴ The pump beam is, in addition, AF modulated, using a rotating sector mechanical chopper on a synchronous motor driven by an audio oscillator and amplifier. A reference signal for the later synchronous AF demodulation is derived from a photodiode viewing a helium-neon laser also chopped by the rotating sector. An alternative would be to audio modulate the RF driving the pump beam electro-optical modulator, using the amateur radio transmitter in AM mode.

The modulated pump and probe beams are focussed into the sample which acts as a "molecular mixer" in which the photons from the pump beam affect the sample (for example by exciting an electronic transition or by starting a chemical reaction) in such a way that the absorption or scattering of the probe beam is altered. Thus the RF frequencies of the pump and probe beams are mixed by the molecular interaction to produce sum and difference frequencies. Detection is by a photo-diode (EG & G Model DT 110) which is directly connected to the antenna input of a communications receiver (Drake Model R-7). A low pass filter (Allen Avionics Model VFL6P5) between detector and receiver is sometimes helpful. The receiver is set to the sum or difference frequency, and thus rejects the RF frequencies of both the pump and probe beams with the sharp discrimination characteristic of a good communications receiver in which filtering is carried out at a fixed intermediate frequency (IF). The

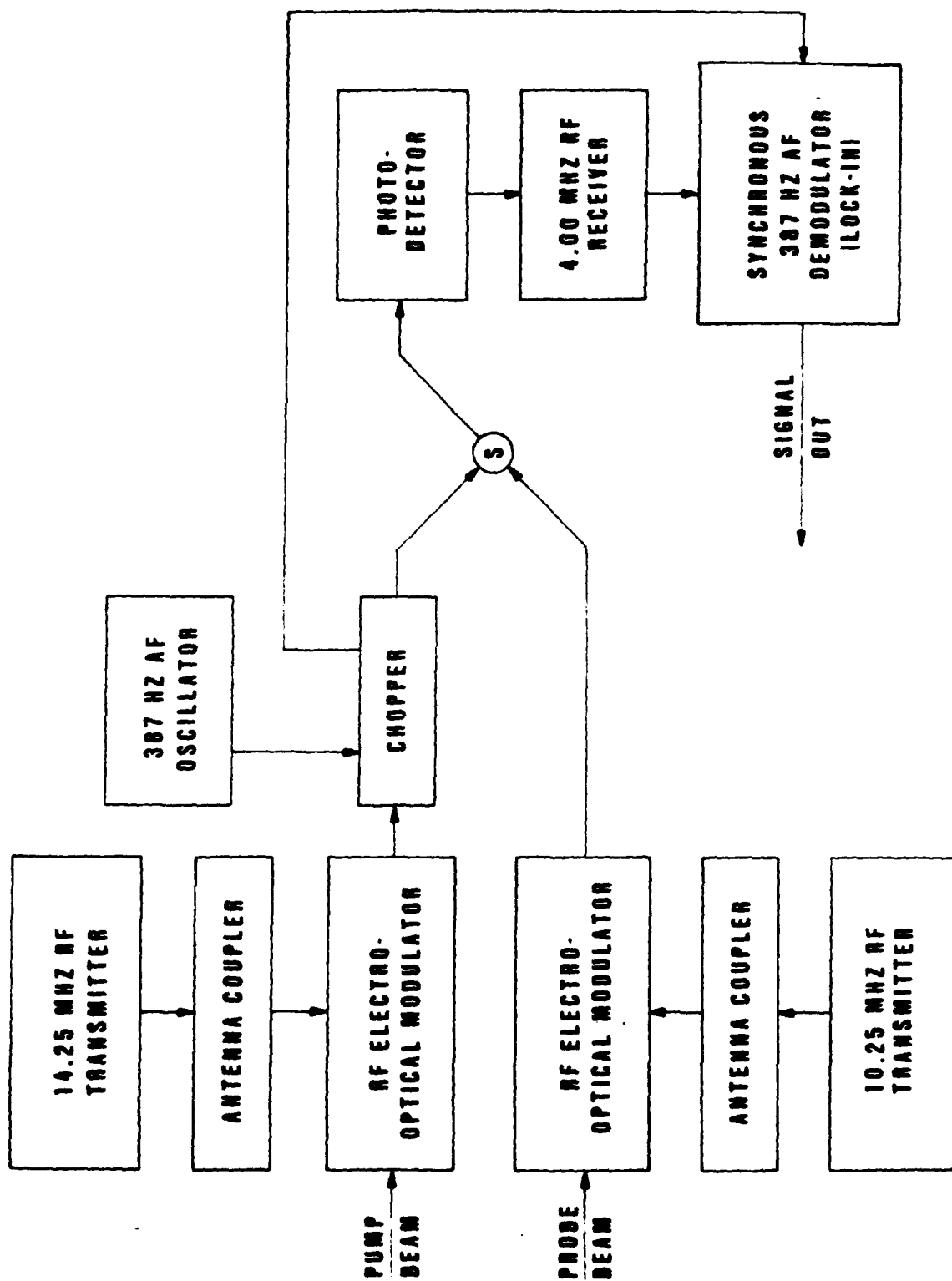


Fig. 1. Block diagram of multiple modulation and detection system.

automatic gain control of the receiver is turned off to maintain linearity, AM detection is used, and the receiver headphone output is fed into an AF synchronous demodulator ("lock-in detector") (Princeton Applied Research Model 124 with Model 116 preamplifier). The reference signal for the demodulator is derived from the AF modulation of the pump beam. The commercial "lock-in detector" could be replaced with a simple phase-locked loop synchronous demodulator⁵ at a considerable savings in cost.

Given the success of frequency modulation (FM) in high-fidelity, low noise, commercial broadcasting, it might be thought that FM of the pump RF drive followed by FM instead of AM detection in the receiver might be advantageous. Unfortunately, under the low signal-to-noise conditions in which multiple modulation is needed, the advantages of FM disappear.⁶ A superior alternative⁷ would be to forego the AF modulation and to lock the local oscillators of two identical receivers together: receiver i) being fed by the detector viewing the "molecular mixer" and receiver ii) by an ordinary mixer generating the sum or difference of the two RF modulation frequencies. The output of the last IF stage of receiver i) (50 kHz for the Drake R-7) could then be synchronously demodulated using the output of the last IF stage of receiver ii) for example with a "lock-in detector".

The rms noise background, measured through calibrating the gain of the detection components, is close to the rms noise theoretically expected⁸ from randomly arriving photons (shot noise) and synchronous demodulation. The measurement was made using 3.6×10^{-2} W of 514.5 nm light from a mode-locked Argon ion laser (certainly not a white noise source) which produced $\sim 3 \times 10^{-11}$ W of rms noise with the synchronous demodulator running into a low pass filter with a 10 second time constant (2.5×10^{-2} Hz equivalent noise bandwidth at 6 dB/octave roll-off). The RF carriers of both the pump and probe beams are thus rejected, even with minimal shielding of the transmitters and with full illumination by the probe beam along with large scattering of the pump beam into the detector. This multiple modulation scheme, which combines the simplicity and high performance of low frequency synchronous detection with the low background noise of RF frequencies^{2,3} should be applicable to a variety of infrared, Raman (resonance and non-resonance), and electronic pump and probe experiments, as well as to auto- and cross-correlation measurements of pulse shapes in the time domain. It is particularly advantageous when pump and probe wavelengths are the same and optical spectral blocking of the pump beam is ruled out.

We thank Dr. J. P. Heritage for helpful discussions, and the National Science Foundation, Chemistry and the Office of Naval Research, Chemistry for the support which has made this work possible.

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