# stepladder to the laser

The ladder which Dr. Oka produced was in the form of radiation from a microwave source. This radiation is of lower frequency than the infrared laser radiation and may readily be produced in a tuneable form using conventional apparatus. By adding, or subtracting, microwave radiation to the single laser frequency, Dr. Oka is able to produce a radiation source which possesses all the advantages of laser radiation combined with the versatility of a variable frequency source. In a typical experiment, the two forms of radiation enter an absorption cell containing a sample of the molecules in gaseous state. The microwave apparatus is then tuned until the resultant radiation exactly matches that necessary to cause a molecular transition.

Since all radiation is present in the form of discrete energy units called photons, the technique is known as Two Photon Spectroscopy. For excitation to take place, the molecule must absorb one photon from the laser radiation and one from the microwave radiation. Using this technique, Dr. Oka is able to extend the study of infrared spectra not only by the increased precision of his measurements but in the detection of hitherto unobservable transitions. Indeed the instrument, which was built by Dr. Oka and Postdoctorate Fellow, Dr. S.M. Freund, immediately reached a limit of resolution which appeared to have been set by nature herself, the actual width of the spectral lines.

Transitions which appear as sharp lines in conventional spectroscopy are seen to have a finite width under the keener scan of Dr. Oka's instrument. Since the Double Photon Spectroscope is capable of readings which are several orders of magnitude finer than the width of these spectral "lines", it becomes important to overcome this problem of uncertainty in the frequency of transitions. The problem of line widths of the spectrum and its major cause is known as Doppler broadening. The solution, involving what is known as the Lamb dip method, required Dr. Oka to further modify his apparatus.

Early in the last century, the physicist Christian Johann Doppler pointed out that the frequency of the radiation which a moving body experiences depends upon that body's velocity relative to the source of radiation. A familiar example of this Doppler shift in frequency is the change in perceived pitch of a train whistle as the engine approaches and then recedes. A similar effect is present in spectroscopic experiments since the molecules of gas under study are in constant chaotic motion, each of them experiencing a slightly different frequency of radiation. The resultant effect is the production of an absorption spectrum in which absorption occurs over a band of frequencies rather than in a sharp line. The modification, which enables Dr. Oka to overcome this smearing of frequency, is to cause radiation to hit the molecules from two opposite directions. In such an arrangement, the moving molecule experiences an increase in frequency in the radiation coming from one direction and a decrease in frequency in the radiation arriving from the opposite direction. The net effect is a cancellation of signal except in those cases for which the molecule is experiencing no Doppler shift. This Lamb dip technique further increases the accuracy of Dr. Oka's measurements and, he says, "the combination of Two Photon Spectroscopy with the Lamb dip has improved the accuracy of infrared spectroscopy over a hundred times and, if we try harder, then much, much more."

Using these techniques, Dr. Oka has made an extensive study of the infrared spectrum of ammonia ($NH_3$), methyl fluoride ($CH_3F$) and phosphine ($PH_3$), giving accurate information of their vibrational behavior. In addition to this refinement of data, he has been able to make observations of previously unobserved transitions. There are transitions which are not observed in normal single photon spectroscopy because they are forbidden by the constraints of molecular symmetry. Absorption of two photons during an excitation process, however, enables this prohibition to be overcome, providing new information on molecular excitations.

**A double photon transition between two vibrational energy levels of a molecule. In the first case (a) the photon from the laser, $\nu$ laser, is unsufficient to cause a transition and requires the aid of a photon from the microwave radiation, $\nu$ microwave. In the second case (b) the laser frequency is too large and the microwave radiation acts in a subtractive manner to induce the transition.**

**Une transition à double photon entre deux niveaux énergétiques vibrationnels d'une molécule. Dans le premier cas (a) le photon émanant du laser, $\nu$ laser, est insuffisant pour déclencher la transition et il est nécessaire d'ajouter un photon émanant du rayonnement en micro-ondes, $\nu$ micro-onde. Dans le deuxième cas (b) la fréquence du laser est trop grande et le rayonnement en micro-ondes permet de soustraire de façon à déclencher la transition.**

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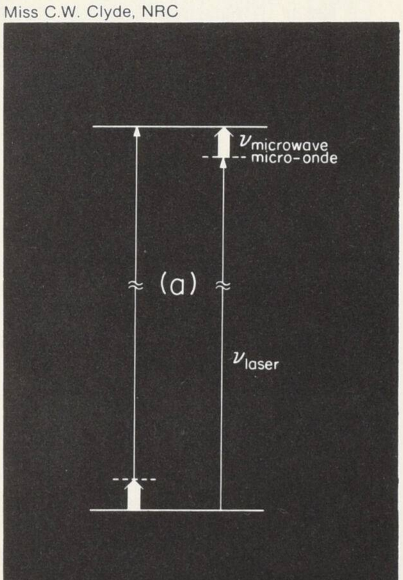


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