

100-200-100-1000

1 of 1

Conf-9301119--3

RECEIVED
OCT 14 1993
OSTI

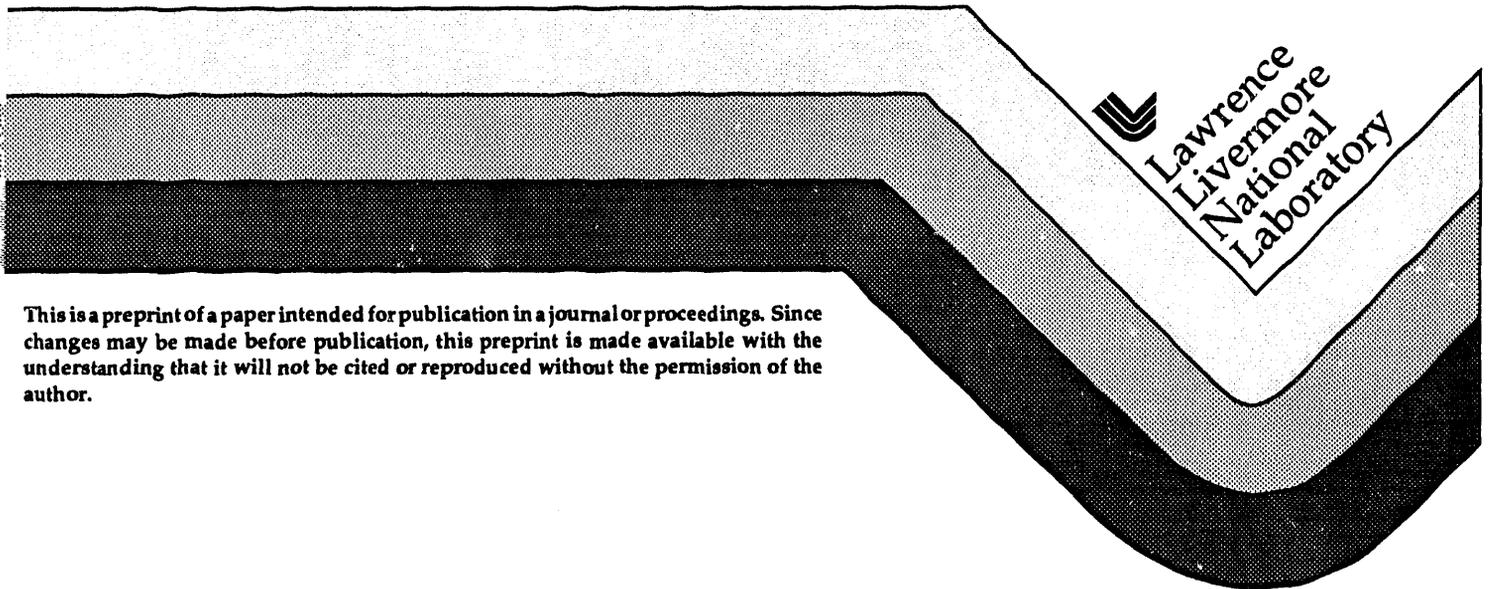
UCRL-JC-114025
PREPRINT

High-Order Sum and Difference-Frequency Generation in Helium

J. K. Crane
M. D. Perry

This paper was prepared for submittal to the
NATO on Super Intense Laser Atom Physics
Han-sur-lesse, Belgium
January 8-14, 1993

May 12, 1993



This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

878

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial products, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

HIGH-ORDER SUM AND DIFFERENCE-FREQUENCY GENERATION IN HELIUM

John K. Crane and Michael D. Perry

Laser Program, P. O. Box 808, L-493
Lawrence Livermore National Laboratory
Livermore, California 94550

INTRODUCTION

High-order harmonic generation provides a new method for generating coherent, XUV radiation.^{1,2} These harmonics are characterized by a rapid, perturbative drop at low orders, followed by a broad plateau extending to photon energies of 150 eV in the lighter, rare gas atoms.^{3,4} An experimentally observed limit coincides with the theoretical limit for harmonic generation in neutral atoms given by the expression $E_c(\text{eV}) = IP(0) + 3U_p(I)$ ⁵, where E_c is the energy cutoff of the harmonic plateau, $IP(0)$ is the field-free ionization potential and U_p is the electron quiver energy at the maximum intensity, I , seen by the atom. As part of a broad effort to develop this technique into a general purpose, XUV source, extensive work to understand the phase-matching between the harmonic and driving fields, and the resulting effect on the conversion efficiency, angular distribution and spectral brightness has been undertaken at several laboratories.⁶⁻⁸ Nevertheless, certain aspects of the harmonically generated radiation such as the polarization, relative strength of a given harmonic, and the plateau extent, are defined by the single atom-field interaction. Specifically, the single-atom harmonic spectrum is determined primarily by the interaction of a driven, quasi-free electron with the atomic potential. Using two, independent fields one can

affect the electron motion by controlling the relative strength, polarization, and phase of the fields and alter the harmonic spectrum.

In this paper we discuss initial, two-color experiments where we drive the atom with two fields of different, but related frequencies: 1053 nm (1ω) and 526 nm (2ω). In addition to the high-order, odd harmonics, we observe sets of three additional peaks that we attribute to sum and difference-frequency generation between the two fields. By controlling the relative polarization between the two fields we can control the relative strength of the harmonic and mixing components, as well as the polarization of the output XUV photon.

Previous work on the interaction between intense, two-color radiation and atoms has concentrated on the influence of the relative phase of the fields on the ionization rate and electron spectrum. Muller et al.⁹ investigated above-threshold ionization (ATI) in krypton produced from two-color illumination by intense (10^{13} W/cm²) 1064 and 532 nm laser pulses. They observed some additional structure in the ATI spectrum when the two fields were present with their relative polarization parallel. Several groups have examined^{10,11} multiphoton ionization using two-color excitation and have shown that the relative phase between the two fields affects the ionization rate. Schafer and Kulander¹⁰ also described calculated harmonic spectra from two-color illumination that contain both odd and even orders. In addition, much attention has been focused recently on manipulating chemical reactions and molecular dynamics by controlling the phase of the coherent field driving the interaction¹².

DESCRIPTION OF THE EXPERIMENT

Our laser is a neodymium:glass system¹³ that can deliver up to 8 Joules of 1053 nm radiation in a 800 femtosecond pulse. The laser is sent into a vacuum chamber, as shown in figure 1, and focused immediately below the output of a high-pressure (up to 1500 psi) pulsed valve equipped with a supersonic (Mach 8) nozzle that can produce densities in excess of 10^{19} atoms/cm³.¹⁴ Light created in the interaction region is collected by a grazing incidence, x-ray spectrometer and imaged onto the input photocathode of an x-ray streak camera. The spectrometer-streak camera combination produces time-resolved, single-shot spectra covering a 20 nm range, allowing us to separate the high-order harmonics from the spontaneous emission produced in the plasma¹⁵

We frequency double the 1053 nm laser light (1ω) with a thin (4 mm) KD*P crystal that is cut for type I phase matching. Although the laser can produce up to 4 Joules of frequency-doubled light, we operated with 1.5 Joules of 1053 nm light input to produce 300 mJ output at 526 nm (2ω). The laser light is focused into the vacuum chamber with a single element, fused silica, aspheric lens that has a 200 cm focal length at 1053 nm. Chromatic dispersion in the lens causes the 526 nm light to focus 5 cm before the 1053 nm light. In these experiments we placed the gas jet at the focus of the 2ω beam. We operated with 200-300 mJ of 2ω light focused to a 50 μm waist (diameter at $1/e^2$ value in intensity) and a 600 femtosecond pulse width (FWHM), measured with a single-shot autocorrelator. The peak intensity based on these parameters is $5 \times 10^{16} \text{ W/cm}^2$. This value is confirmed by observing fluorescence from Ar VIII transitions which occur only above $3 \times 10^{16} \text{ W/cm}^2$.* The beam diameter of the 1ω light at the gas nozzle is 1.0 mm with a corresponding peak intensity at the jet of $6 \times 10^{13} \text{ W/cm}^2$ in an 800 femtosecond pulse.

A temporal walkoff between the 1ω and 2ω pulses occurs because of the difference in group velocity between the two pulses propagating in the fused silica lens and the KD*P doubling crystal. We use this temporal walkoff as a means of varying the overlap between the two colors. We calculate a walkoff parameter, $d_{12} = (v_g(1\omega))^{-1} - (v_g(2\omega))^{-1} = 0.79 \text{ psec/cm}$, in fused silica. The combination of the focusing lens and doubling crystal separate the pulses by approximately 0.8 psec. To further separate the pulses we add a 15 mm optical flat, so that the combined effect of the two pieces of fused silica plus KD*P crystal produces a 2.0 psec time separation between the peak of the two pulses. Figures 2a and 2b shows the temporal relationship between the two pulses for the two different cases. Since the intensity of the 2ω pulse is many times our measured saturation intensity for ionization of neutral helium ($6.5 \times 10^{14} \text{ W/cm}^2$)¹⁶, the harmonics are produced during the rising edge of the 2ω pulse. Although the peak intensity of the 2ω field is $3 \times 10^{16} \text{ W/cm}^2$, essentially all harmonic production from neutral helium will occur below $1 \times 10^{15} \text{ W/cm}^2$. This intensity occurs approximately 0.8 psec before the peak of the 2ω pulse, nearly coincident with the peak of the 1ω pulse with only the group delay of the lens and the KD*P present. When the extra 15 mm fused silica optic is added the total delay is 2.0 psec and the 1ω peak coincides with a 2ω

*We calculated the appearance irradiance for Ar VIII lines using the tunneling ionization formalism described by M. V. Ammosov, N. B. Delone, and V. P. Krainov, *Sov. Phys. JETP* . 64:1191 (1986).

intensity less than 1×10^{12} W/cm², well below the intensity for harmonic production in helium.

4

We use a type I doubling crystal to convert the 1053 nm light to 526 nm. The frequency-doubled field is orthogonally polarized with respect to the fundamental (1ω), however we can rotate the polarization of the 1ω field after doubling using a half-wave plate with little effect on the 2ω polarization. We use the half-wave plate to continuously vary the angle between the polarization of the two colors in our experiments.

RESULTS

Sum and Difference Frequency Generation

We measured the harmonic spectrum at various laser irradiances for the following four cases: (a) 2ω only; (b) 1ω only; (c) $1\omega+2\omega$ without the 15 mm optical flat; and (d) 1ω and 2ω with the optical flat. By measuring the spectrum with and without the optical flat, we could insure that everything was held constant, except the temporal overlap between the pulses. In figure 3 we display the spectra for three of the four cases. Figure 3a shows the harmonic spectrum produced when only 2ω is focused into the chamber (1ω is blocked by a thin absorbing filter placed after the doubling crystal). In this figure, we show the region covering 34 to 69 eV. The 15-27 odd harmonics are observed in first order along with weaker second order peaks at the 29th and 31st harmonics at this spectrometer setting (the spectrum extends up to the 35th harmonic of the 526 nm light). This result is identical to our previously reported results.¹⁶ In the second case, we send only 1ω light into the chamber (detune or remove the doubling crystal) and observe no harmonics. The absence of odd harmonics of 1ω light is not surprising at this intensity (6×10^{13} W/cm²), which is an order of magnitude below the intensity where we normally observe harmonics in this spectral range (29th-55th of 1ω)¹⁶. We do not show this null result. Figure 3b shows the third case where both colors are sent into the chamber, separated by 800 femtoseconds. Again we observe the odd harmonics of 2ω , however, between pairs of 2ω harmonics we observe three additional peaks. This pattern repeats itself throughout the range of observed 2ω odd harmonics, extending to the plateau cutoff. In the final case, shown in figure 3c, we place the additional 15 mm optical flat in the beam, separating the two colors in time by 2.0 psec. The result is identical to figure

3a- odd harmonics of 2ω only. We repeated this sequence of four experiments throughout the spectral range of our spectrometer, spanning the region from the 11th harmonic of 2ω (26 eV) to beyond the cutoff for 2ω harmonics (35th order at 83 eV), observing the same result as that shown in figure 3.

5

Of the three secondary peaks in between the usual odd harmonics of the 526 nm field, we concentrate first on those centered between the odd harmonics. These peaks appear at even multiples of the 526 nm field. Upon initial inspection, we would label these as the 18th, 20th, 22nd, etc. harmonics of the 2ω field. These *even* harmonics can only be produced by breaking the symmetry of the light-atom Hamiltonian, e.g. with a second field. Alternatively, a sum-frequency mechanism can be used to explain the peaks that appear at even multiples of the 526 nm field. In this case, an odd number of 2ω photons combine with two 1ω photons to produce a single photon with an energy equal to an even multiple of a 2ω photon. This mechanism is fully dipole allowed and the overall harmonic order is still odd. *It is only the energy of the photon which makes it appear as an even harmonic.* Clearly, harmonics produced by this sum frequency mechanism are only produced when both the 1ω and 2ω fields are present.

The secondary peaks on either side of the principal odd harmonics ($q\omega$) of the 2ω field have an energy equal to odd multiples of the 1ω field. However, these are not 1ω harmonics in the usual sense as they only appear when both fields are present. In this case, the emission results from either sum or difference-frequency generation with an *even* number ($q+1$) of 2ω photons. The peak just below the q th odd harmonic is formed by the absorption of $q-1$ photons of the strong 2ω field plus one additional 1ω photon. The peak just above the q th odd harmonic is formed by the absorption of $q+1$ photons minus a single 1ω photon.

Figure 4 is an expansion of the portion of figure 3b that spans the 17th through 21st harmonics of 526 nm with the various peaks labeled as described in the previous text. Higher order processes that are degenerate with the three lowest order schemes are also possible and likely as the intensity of the 1ω field is increased. For example we indicate that the ladder $[q+1](2\omega)-(1\omega)$, which contains 21 photons ($20-2\omega$ photons minus one 1ω photon), is responsible for the peak at 45.9 eV. This scheme is degenerate with $[q-1](2\omega)+3(1\omega)$; both involve 21 photons for the excitation and are dipole allowed. We do not believe these higher order diagrams in the 1ω field are contributing significantly at the 1ω irradiance used here. This is based on the fact that the peak which definitely contains two 1ω photons is significantly lower than the peaks which contain only a single 1ω photon.

Polarizations effects

Figure 5a shows a harmonic spectra for the case where the two driving fields are orthogonal to each other. There appears to be a consistent relationship among the three sets of peaks. The odd harmonics of 2ω are the strongest -an order of magnitude larger than the even harmonic peaks. The two sum and difference-frequency peaks that lie to either side of the even harmonic peaks are 2-5 times weaker than the odd harmonic peaks, but consistently stronger than the even harmonic peaks. In figure 5b we show a similar spectrum for the case that the 1ω polarization was rotated after doubling to align it with the 2ω polarization, i.e. the 1ω and 2ω polarizations are parallel. Now all three sets of peaks are comparable in relative strength. We have also rotated the polarization of the 1ω field to intermediate angles between these two extremes and observe that the relative intensities of the sets of peaks can be controlled by this method. Becker et. al.¹⁷ recently showed that by using circularly polarized light for the two-colors certain sets of peaks can be suppressed completely.

Although it may be possible to determine the polarization of the output XUV photons, it is not a trivial measurement to make using standard optical techniques. However by using some simple diagrams we can show the relationship between the polarization of the output XUV photon and the relative polarization of the 1ω and 2ω fields. For the case where both input fields are linearly polarized along the same axis, there will be no change in the orientation between the ground and final states ($\Delta m=0$, where m are the magnetic sublevels), and the polarization of the output, XUV photon will be linear and parallel to the input polarizations. We can explain the sum and difference-frequency peaks for orthogonally polarized fields by a series of dipole allowed transitions. For the even harmonics we add two 1ω photons to an odd number of 2ω photons as described in the previous section. The orthogonal field (1ω) can be expressed as a linear sum of right and left-hand circularly polarized light using the helicity basis set. In this case a two-photon transition can produce a net change in angular momentum, $\Delta m=0$, where the first photon causes a $\Delta m=\pm 1$ transition and the second photon, $\Delta m=\mp 1$, as shown in figure 6a. This sum-frequency mechanism is fully dipole allowed with two fields of orthogonal linear polarization. The two peaks on either side of the harmonic frequencies (either even or odd) are produced by adding (sum-frequency generation) or subtracting (difference-frequency generation) a single, 1ω photon to an even number of 2ω photons. Since the 1ω field is polarized orthogonally to the 2ω field, the upper state will be a coherent superposition of $\Delta m=\pm 1$ states as shown in figure 6b. This last example suggests that by adding (or subtracting) a circularly polarized, 1ω photon, the

final state will be either $\Delta m = +1$ or -1 with respect to the ground state, and the output XUV photon will be circularly polarized. Therefore, by controlling the polarization of the two input fields it may be possible to produce XUV photons of any desired polarization, as well as control the relative strength of the sum and difference-frequency components.

7

Role of relative intensity, phase, and polarization on plateau extension

In the tunnelling limit where $\gamma^2 = IP/2U_p \ll 1$, the harmonic spectra is primarily the result of the field-driven, quasi-free electron interacting with the ion core potential;¹⁸ consequently, high-order harmonic generation, as well as ATI, can be described classically.^{18,19} In this approach, an ensemble of electrons is launched at phases of the laser field determined by the probability for ionization. The trajectory of the electron then determines the probability for harmonic emission by scattering from the core potential. These models are successful at predicting the energy spectrum and cutoff of high-order harmonic generation and ATI when a single field drives the electron. To see how the electron dynamics is affected by two independent fields we derived expressions for the velocity, displacement, and instantaneous kinetic energy starting from the Lorentz force equation. We consider the two simplest cases of parallel and perpendicular polarization for the two fields and write the equations for the instantaneous kinetic energy, $T(t)$. For 1ω perpendicular to 2ω ,

$$T(t) = 2U_p(\omega)\{\sin\omega t - \sin\omega t_0\}^2 + 2U_p(2\omega)\{\sin(2\omega t + \phi) - \sin(2\omega t_0 + \phi)\}^2, \quad (1)$$

and for 1ω parallel to 2ω ,

$$T(t) = 2U_p(\omega)\{\sin\omega t - \sin\omega t_0\}^2 + 2U_p(2\omega)\{\sin(2\omega t + \phi) - \sin(2\omega t_0 + \phi)\}^2 + 2U_p(\omega)[I(2\omega)/I(1\omega)]^{1/2}\{\sin\omega t - \sin\omega t_0\}\{\sin(2\omega t + \phi) - \sin(2\omega t_0 + \phi)\}. \quad (2)$$

In these expressions U_p is the time-averaged quiver energy evaluated at the laser intensity, I , ϕ is the relative phase between the two fields, and t_0 is initial time at which the electron is born. Note the additional interference term when the fields are parallel polarized. Consider an example where the 1ω and 2ω fields are of equal intensity, corresponding to the saturation field strength in helium for a 1 ps pulse ($I(1\omega) = I(2\omega) = 1.6 \times 10^{14}$ W/cm² for $1\omega \parallel 2\omega$) and ($I(1\omega) = I(2\omega) = 3.2 \times 10^{14}$ W/cm² for $1\omega \perp 2\omega$). For an electron born at the peak intensity, i.e. $t_0 = 0$, the quiver energy ranges from 66 to 196 eV for orthogonal polarization, depending

on the phase angle. For parallel polarization the maximum quiver energy range is 48 to 58 eV. In both cases, the harmonic cutoff is pushed beyond that which would be obtained from either field independently at these intensities. However, when compared to the cutoff of the single, long wavelength (1ω) field at the saturation strength (6.4×10^{14} W/cm²), the extension of the cutoff associated with mixed fields is not significant.

8

CONCLUSIONS

We report the observation of high-order, sum and difference-frequency generation in helium. We produce these high-order frequencies as well as the usual odd harmonics by mixing two-colors, 526 nm and 1053 nm, in the high density output of a pulsed supersonic nozzle. What appear as even harmonics can be explained by sum-frequency generation between the two fields. The relative strength and polarization of these different, XUV mixing products is dependent on the relative polarization between the two fields yielding a form of coherent control. As shown in the two-color work on multiphoton ionization⁹⁻¹¹, further control may be possible via the relative phase and strength of the two fields.

We gratefully acknowledge useful discussions with H. T. Powell, J. A. Paisner, S. Dixit, K. C. Kulander, K. Schafer, and T. Ditmire. We would have been unable to perform these experiments without the technical assistance of S. Herman, H. Nguyen, and B. Adams. Work performed under the auspices of the U.S. DOE by LLNL under contract no. W-7405-Eng-48.

REFERENCES

1. A. McPherson, G. Gibson, H. Jara, U. Johann, T. S. Luk, I. McIntyre, K. Boyer and C. K. Rhodes, Studies of multiphoton production of vacuum-ultraviolet radiation in the rare gases, *J. Opt. Soc. Am. B* . 4:595 (1987).
2. M. Ferray, A. L'Huillier, X. F. Li, L. A. Lompre, G. Mainfray, and C. Manus, Multiple-harmonic conversion of 1064 nm radiation in rare gases, *J. Phys. B* . 21:L31 (1988).
3. J. J. Macklin, J. D. Kmetec, C. L. Gordon, High-order harmonic generation in neon, "Proceedings from the Conference on Quantum Electronics and Laser Science", Anaheim, CA (1992).
4. A. L'Huillier and P. Balcou, High-order harmonic generation in rare gases with a 1 ps 1053 nm laser, *Phys. Rev. Lett.* (to be published).
5. J. L. Krause, K. J. Schafer, and K. C. Kulander, High-order harmonic generation from atoms and ions in the high intensity regime, *Phys. Rev. Lett.* 68:3535 (1992).
6. A. L'Huillier, L. A. Lompre, G. Mainfray, and C. Manus, High-order harmonic generation in rare gases, in: "Atoms in Intense Fields," M. Gavrilu, ed. Academic, Orlando (1992).
7. D. D. Meyerhofer and J. Peatross, Angular distributions of high order harmonics, in: "Proceedings on SILAP III," B. Piraux, ed. Plenum, New York (1993).
8. R. A. Smith, J. W. G. Tisch, M. S. N. Ciarrocca, S. Augst, and M. H. R. Hutchinson, Space-resolved ultra-high harmonic generation with picosecond pulses, in: "Proceedings on SILAP III," B. Piraux, ed. Plenum, New York (1993).
9. H. G. Muller, P. H. Bucksbaum, D. W. Schumacher and A. Zavriyev, Above-threshold ionisation iwth a two-colour laser field, *J. Phys. B* . 23:2761 (1990).
10. K. J. Schafer and K. C. Kulander, Phase-dependent effects in multiphoton ionization induced by a laser field and its second harmonic, *Phys. Rev. A* . 45:8026 (1992).
11. R. M. Potvliege and P. H. G. Smith, Two-colour multiphoton ionization of hydrogen by an intense laser field and one of its harmonics, *J. Phys. B* . 25:2501 (1992).
12. M. Shapiro, J. W. Hepburn, and P. Brumer, Simplified laser control of unimolecular reactions: simultaneous (ω_1, ω_3) excitation, *Chem. Phys. Lett.* 149:51 (1988).
13. F. G. Patterson, M. D. Perry, and J. T. Hunt, Design and performance of a multiterawatt, subpicosecond neodymium:glass laser, *J. Opt. Soc. Am. B* . 8:2384 (1991).

14. M. D. Perry, C. Darrow, C. Coverdale, and J. K. Crane, Measurement of the local electron density by means of stimulated Raman scattering in a laser-produced gas jet plasma, *Opt. Lett.* 17:523 (1992).

15. J. K. Crane, M. D. Perry, S. Herman, and R. W. Falcone, High-field harmonic generation in helium, *Opt. Lett.* 17:1256 (1992).

16. J. K. Crane, M. D. Perry, D. Strickland, S. M. Herman, and R. W. Falcone, Coherent and incoherent XUV emission in helium and neon, laser-driven plasmas, *IEEE Trans. Plasma Science* (to be published Feb. 1993).

17. W. Becker, S. Long, and McIver, Two-color higher harmonic production in a zero-range potential, *in*: "Proceedings on SILAP III," B. Piraux, ed. Plenum, New York (1993).

18. P. B. Corkum, et. al., "Short Wavelength V: Physics with Intense Laser Pulses", OSA, Washington D.C. (1993)

19. K. C. Kulander, Dynamics of short-pulse excitation, ionization and harmonic generation, *in*: "Proceedings on SILAP III," B. Piraux, ed. Plenum, New York (1993).

FIGURE CAPTIONS

11

Figure 1. Experimental apparatus for producing and detecting short-pulse XUV radiation.

Figure 2. a) Temporal relationship between 1ω and 2ω pulses where the group delay is produced by the lens and KD*P crystal. b) Additional delay produced by 15 mm thick fused silica optical flat.

Figure 3. a) Harmonic spectrum with 2ω field only shows the odd harmonics 15-27. b) Spectrum from two-color excitation, where the 1ω and 2ω fields are orthogonally polarized and separated in time by 800 fs as shown in figure 2a. c) Spectrum with two-colors separated in time by 2.0 ps as shown in figure 2b.

Figure 4. Enlargement of a portion of the spectrum from figure 3b showing the sum and difference-frequency peaks labelled according to the description given in the text.

Figure 5. a) Spectrum of harmonic and sum and difference-frequency peaks where the polarizations of the 1ω and 2ω driving fields are perpendicular. b) Spectrum generated by the two-colors polarized parallel with respect to each other.

Figure 6. a) Excitation ladder for two, orthogonally polarized fields: odd number of 2ω photons plus 2- 1ω photons. b) Excitation ladder for orthogonally polarized fields: even number of 2ω photons minus a single, 1ω photon.

Vacuum chamber and optics used in harmonic generation experiments

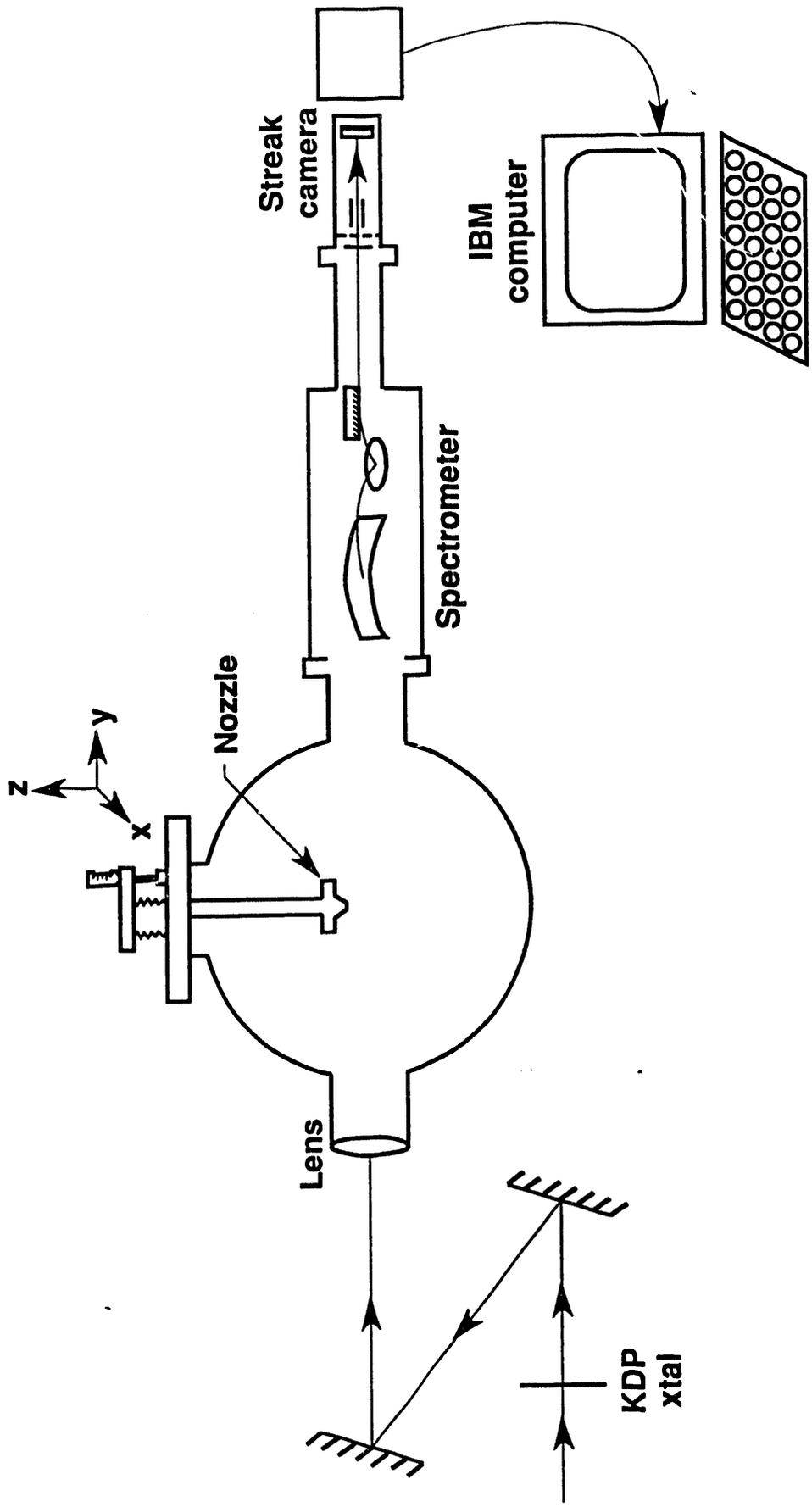


Fig. 1

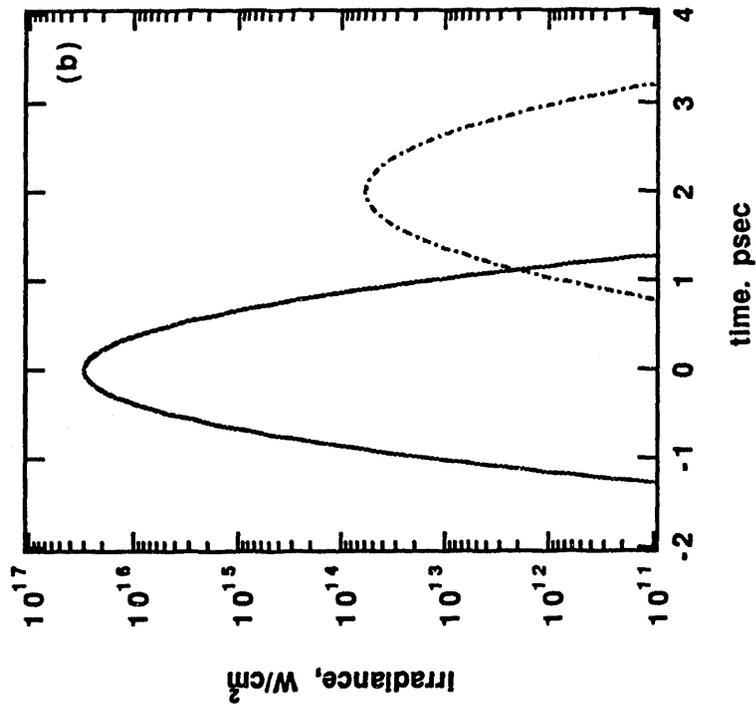
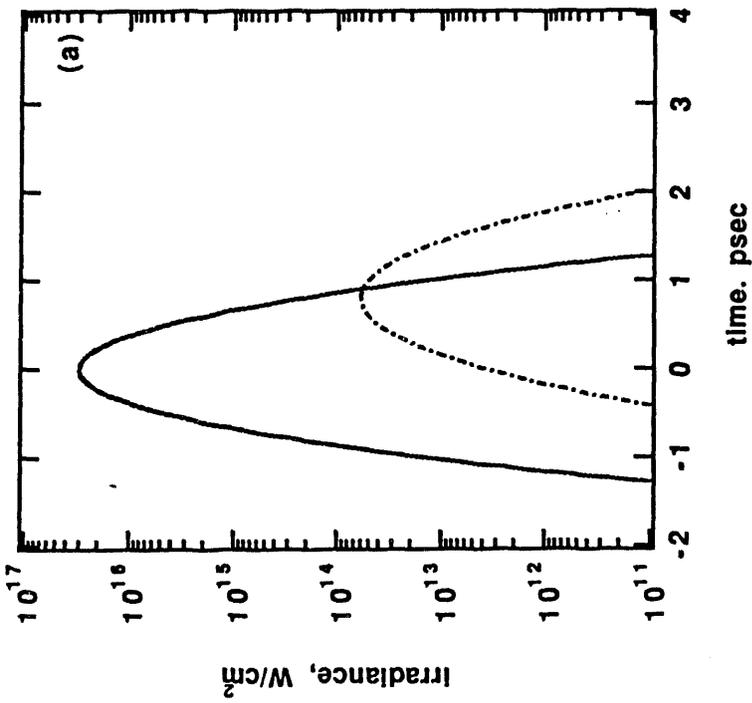


Fig 2a

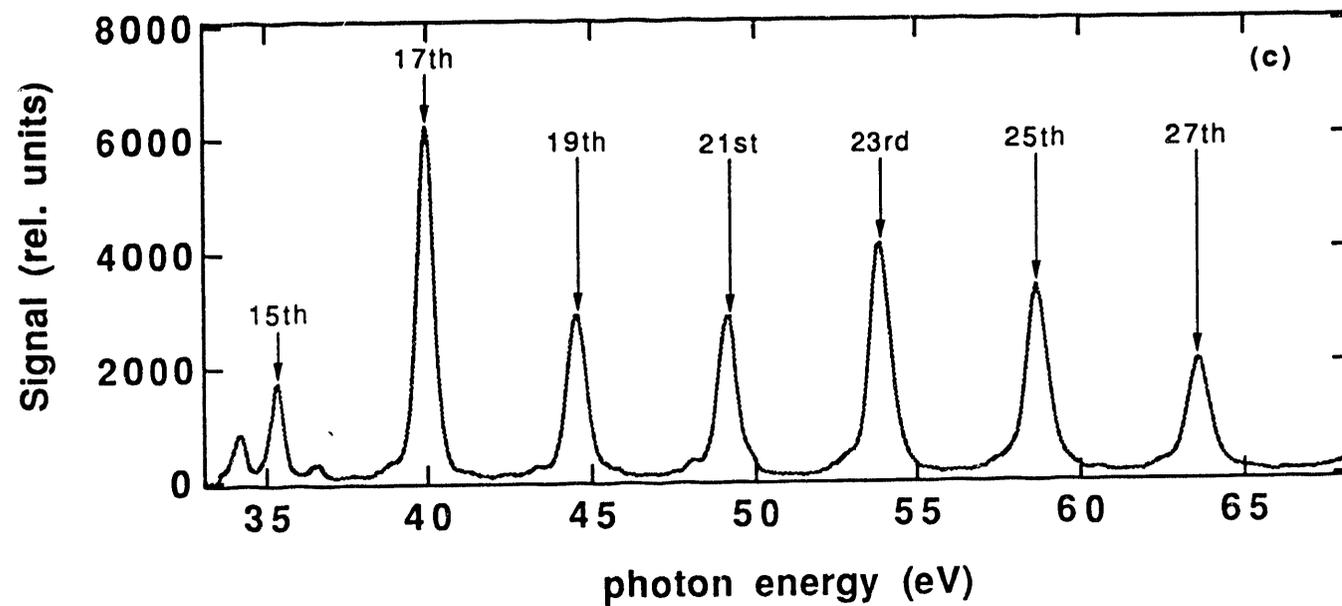
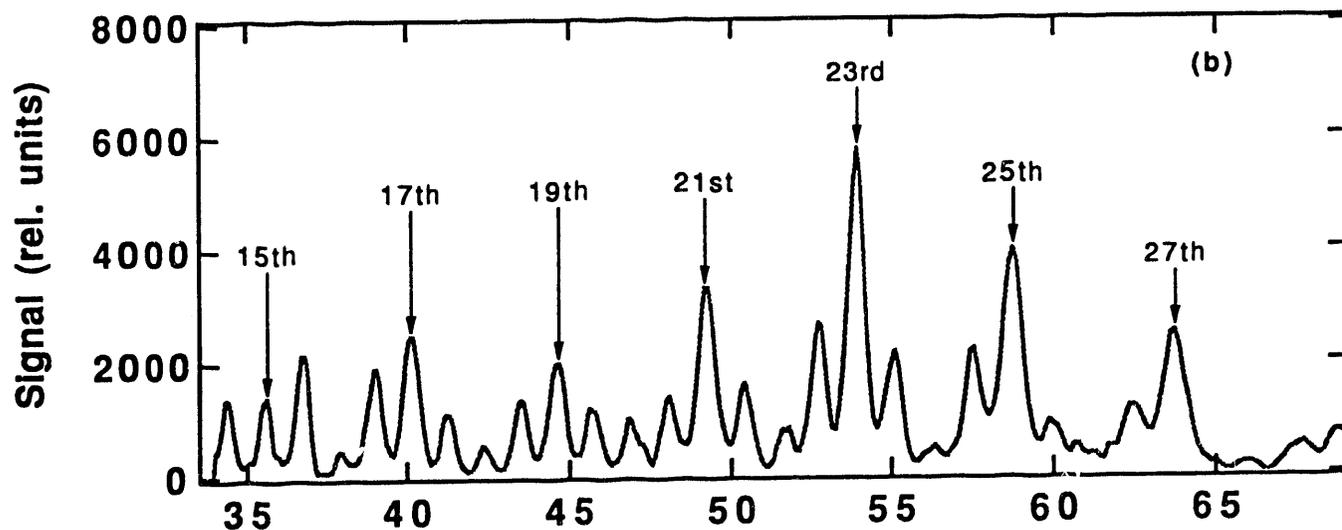
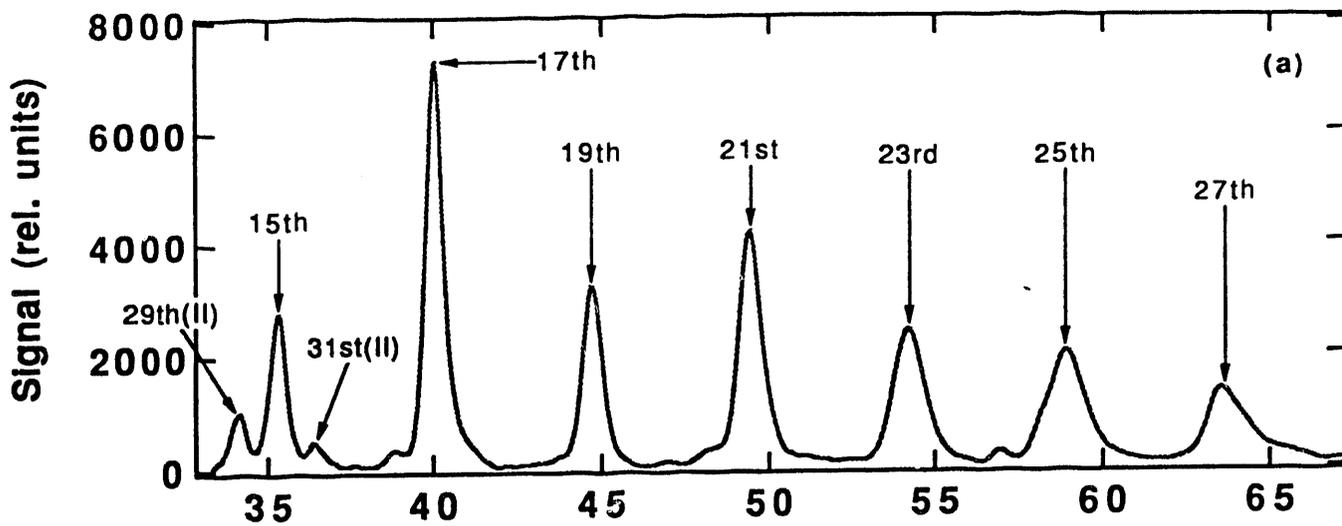


FIG 3

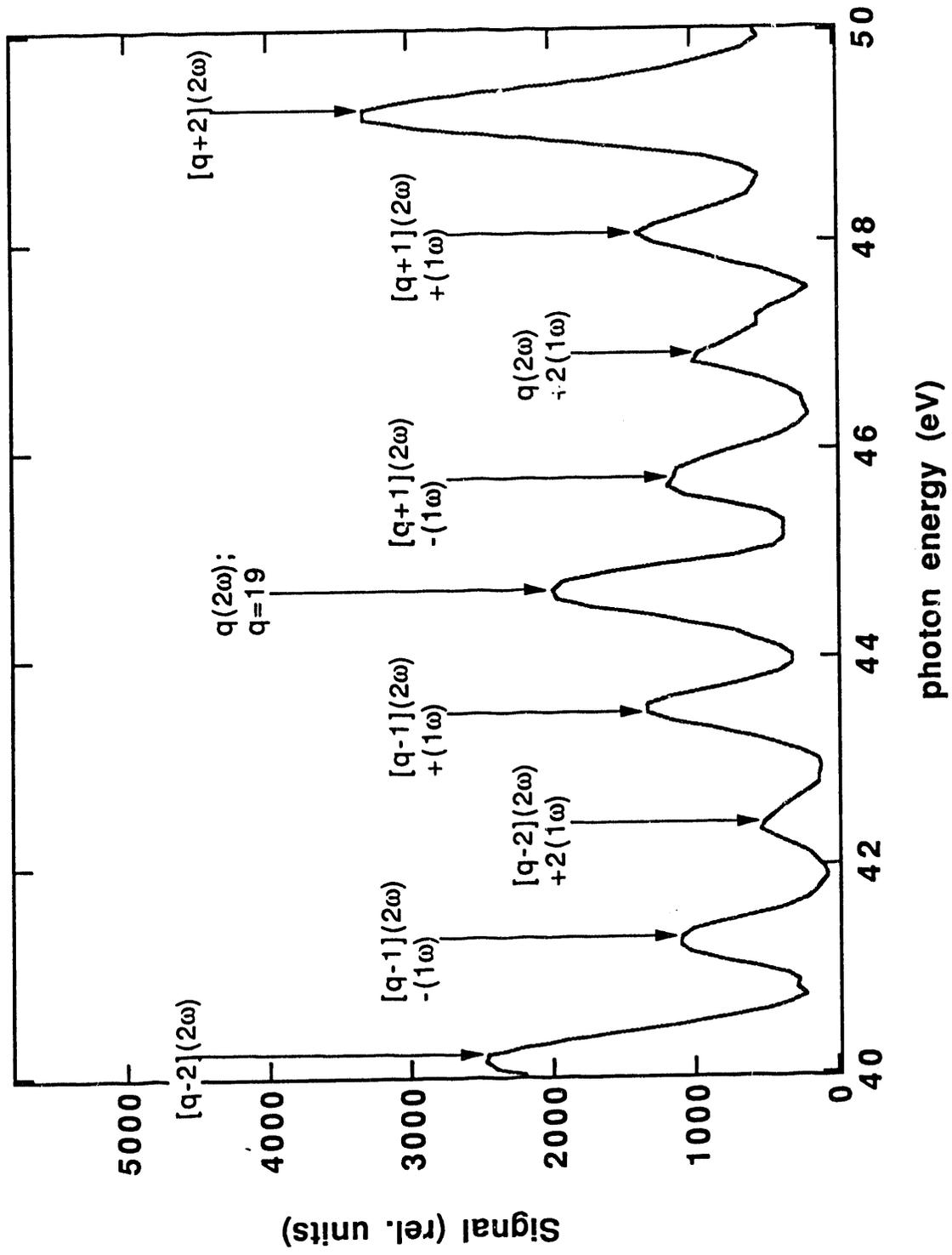


Fig. 4

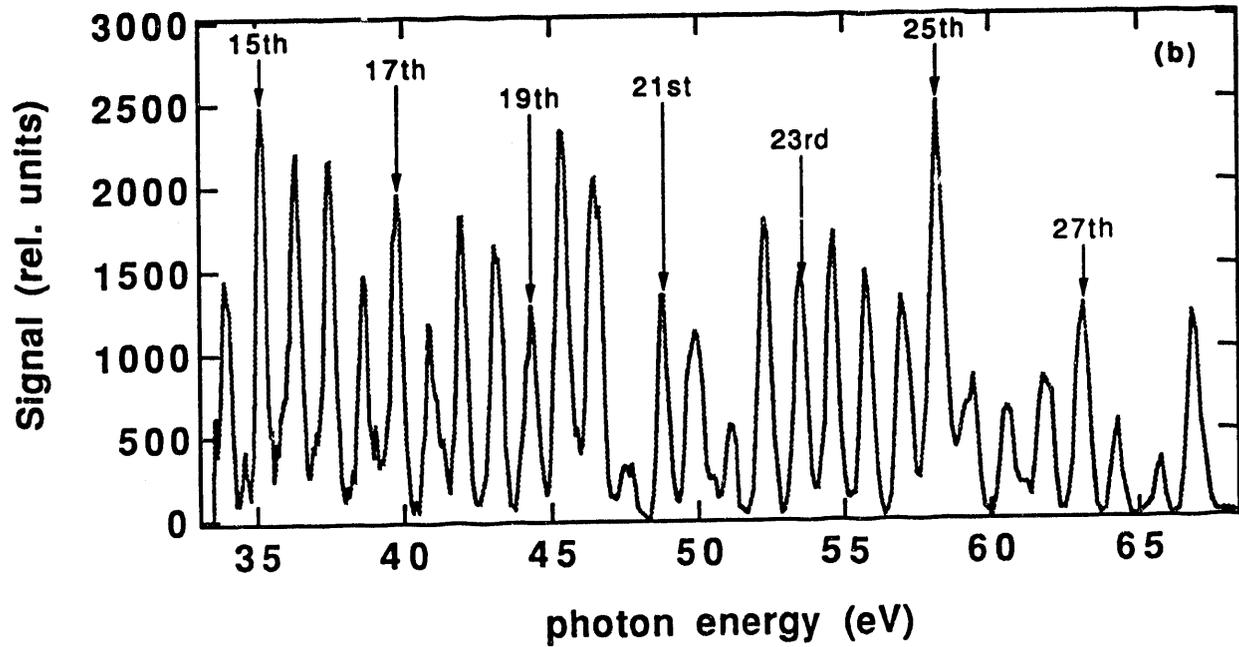
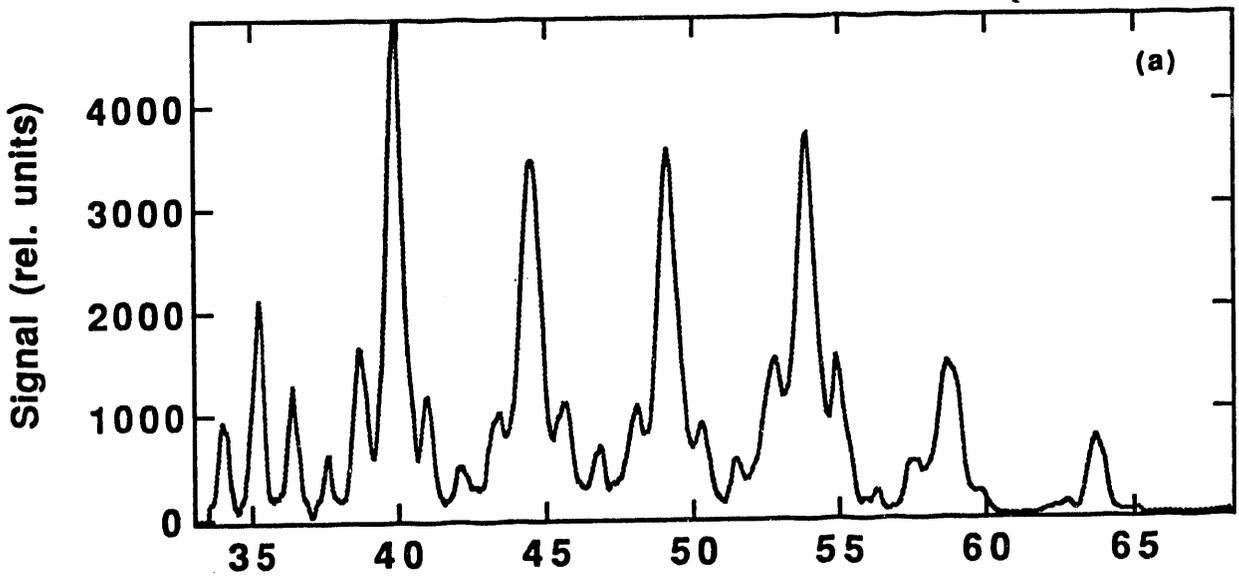
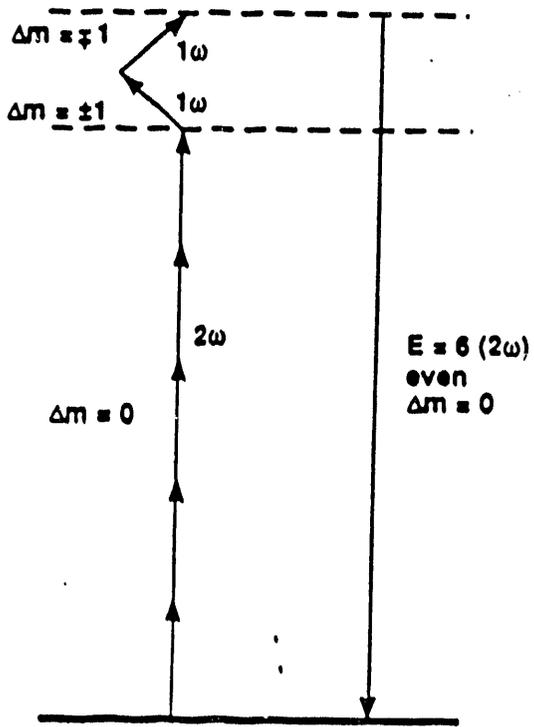
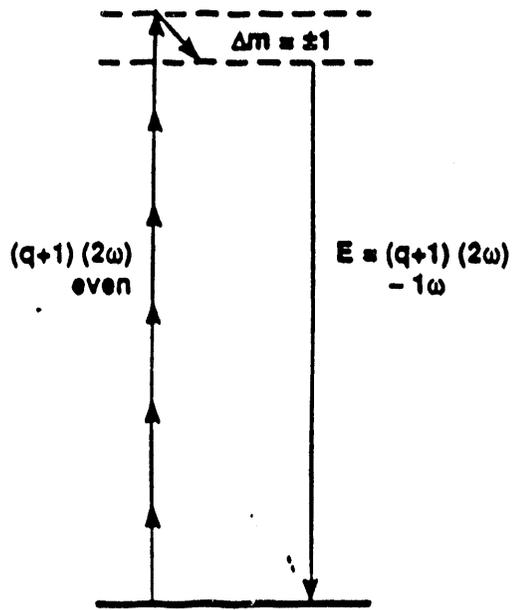


Fig. 5



(a)



(b)

Fig 6

DATE

FILMED

12 / 14 / 93

END

