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Extreme Broadening of Stimulated Raman Scattered Light From High Intensity Laser Plasma Interactions

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High intensity picosecond laser plasma interaction experiments were performed to examine nonlinear scattering mechanisms in field ionized underdense plasmas. Broad and oscillatory spectra were observed for the forward scattered light. The Raman backscattered spectrum showed an extremely broad, supercontinuum like nature, extending from 450 nm to greater than 1200 nm at incident laser intensities of 2 X 10^{18} W/cm^2. Narrow and large amplitude modulations in the spectrum of the backscattered radiation were measured and are attributed to scattering from ion waves.

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EXTREME BROADENING OF STIMULATED RAMAN SCATTERED LIGHT FROM HIGH INTENSITY LASER PLASMA INTERACTIONS

High intensity short pulse laser systems have become widely available in the past few years largely due to the commercialization of CPA type laser designs [1]. Such lasers have potential use in a wide variety of applications ranging from the Laser Wakefield Accelerator [2], and x-ray lasers [3] to Inertial Confinement Fusion experiments [4]. However, before such applications can be realized, the basic physics of very high intensity laser plasma interactions must be examined experimentally to determine the validity of theoretically predicted phenomena and if there remain issues which have yet to be considered. The detection of scattered laser light has particular relevance for diagnosis of laser produced plasmas since by such measurements it is possible to provide conclusive evidence for phenomena such as the production of plasma wakefields (for electron accelerator applications) as well as to accurately determine fluctuation levels and the plasma electron density.

For extremely high intensity laser light, the motion imparted to the electrons can become relativistic and many new effects associated with the increasing mass of the electron have been predicted [5]. A measure of the importance of relativistic effects in an interaction is given by the parameter $a_o = qE_{laser}/cm_e\omega_o$ where ω_o is the laser frequency (note that a_o is also the nonrelativistic quiver velocity, v_{osc}/c, of the electrons in the laser field). In particular, the mechanisms for Thomson scattering [6] and stimulated Raman scattering (SRS) [7] become highly nonlinear and emission of scattered radiation at harmonics of the laser frequency have been predicted. In addition, the intense ponderomotive forces created by high intensity picosecond laser pulses excite large amplitude plasma waves which in turn self-modulate the laser pulse. In addition, scattering of laser light by collective processes like SRS is important in laser-plasma interactions since it can be a principle source of electron heating and laser absorption.

Experiments to examine high intensity SRS were undertaken at the Naval Research Laboratory using the table top terawatt laser facility which produces pulses at a wavelength of 1.054 μm having a maximum energy of 1.2 J and a temporal width of about 800 fsec. The beam has a "top hat"profile and is measured to be 1.4 times diffraction limited. This laser has a

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maximum focused intensity of 2.1×10^{18} W/cm^2 at $\lambda = 1.054$ µm which gives a value for $a_o \sim 1.1$ indicating that effects due to relativistic phenomena may be important in these experiments. The focal spot diameter of the laser pulse in vacuum was measured to be 8.5 µm although it was not measured during interaction with the gas jet target. All experimental intensity values presented here are understood to imply the expected vacuum intensity values without taking into account self-focusing or defocusing effects in the interaction region.

The optical setup for these experiments was situated close to the final laser table in order to minimize the value of the B integral (self-phase modulation) by limiting the length of air through which the full power pulse travels ($\approx$ 3 m). The laser beam was focused by an f/3 off-axis parabolic mirror into a pulsed gas jet which is capable of producing atomic densities of up to 5×10^{18} cm^{-3} (n_e up to 10^{19} cm^{-3}) with plenum pressures of 11 atmospheres. Both helium and hydrogen were used as target gases in these experiments. At relatively low intensity (up to 10^{16} W/cm^2) the light directly backscattered by stimulated Raman scattering (SRS) shows a single narrow peak - frequency downshifted by the electron plasma frequency. The amount of this shift allowed a precise measurement of the plasma electron density.

The breakdown of the gas by the laser pulse was recorded by a CCD camera positioned at a 90° observation angle to the direction of the incident laser. The characteristics of the breakdown were highly reproducible - however at maximum target gas pressures and highest laser intensities it was noticed that the region of laser breakdown usually included several separate foci. This has been observed previously [8] and such behavior was interpreted as due to relativistic self-focusing.

In general, the forward scattered light showed a large spectral broadening - up to 150 Å - (see Fig. 1) in addition to a blueshift of up to 50 Å, (although through the use of a preionizing pulse the average blueshift could be reduced to less than 10 Å). This blueshift is dependent on the rate of ionization of the gas by the focused laser light and has been observed previously[9]. The broadened spectrum exhibits an oscillatory structure extending into both red and blue regions surrounding the fundamental. Similar structures in forward scattered spectra have been observed previously and were associated with supercontinuum generation [10]. The oscillatory behavior measured here is not a consistent one and the detailed features of the spectrum are not reproducible from shot to shot although the modulation intensity and periodicity is approximately constant. Anti-Stokes forward Raman scattered emission was also observed. Total absorption and scattering of light by all processes at $I = 2 \times 10^{18}$ W/cm^2 and $n_e = 6 \times 10^{18}$ cm^{-3} was not more than 10 - 15 %.

In order to determine the high intensity behavior of the SRS instability, backscattered light was examined through the use of two spectrometers (Minuteman 305M and Jarrell Ash Model 1233) which were operational in the wavelength range 300 - 1200 nm. A CCD array was used as a detector. The backscattered light was transmitted through a fused silica turning mirror which was dielectrically coated to reflect in the region 1000 - 1100 nm. This turning mirror acted as a 10^{-3} filter in this wavelength region although backscattered light in this range was strong enough so that light which leaked through was also measurable. As the laser intensity was increased up to 2×10^{18} W/cm^2 (see Fig. 2) the backscattered Raman peak was blueshifted and became much broader until it extended from about 450 nm to greater than 1200 nm (which was the upper detectable limit of the spectrometers used here). No spectrometer available to us had a spectral range large enough to encompass the entire backscattered spectrum on a single shot. The amount of broadening of the SRS emission observed here was much larger than broadening of backscattered Raman emission observed previously [11]. In fact, previous experiments (in which the target gas was confined in a cell) showed a sharp Raman peak (low intensity effect) superimposed upon a broad slightly blueshifted low level signal - which was interpreted to be the effect of the highest laser field regions. Here, no sharp peak remains at the wavelength of the plasma frequency shift - although the intensity of the scattered light is greatest by several orders of magnitude in the region near the plasma frequency shifted light (somewhat blue shifted as intensity is increased) and falls off gradually to the blue and the red. A slightly blue shifted peak in the backscattered spectrum also begins to develop close to the fundamental frequency as the intensity is increased above 10^{17} W/cm^2. This observed feature increases exponentially with respect to the incident laser intensity .

The most prominent feature of this backscattered spectrum is an oscillatory structure which is superimposed on the broad backscattered signal. As shown in Fig. 3 the oscillatory period was approximately 30 Å in the region of the fundamental and seemed to decrease to about 10 Å as the edge of the broadband signal was approached. In frequency space however the period does not change appreciably. In Fig. 4, several spectra were converted from wavelength to frequency space and were Fourier transformed in order to determine if the oscillations correspond to known frequencies of the plasma.

The following is a discussion of the experimentally observed results. The appearance of multiple foci in these experiments may not be caused by simple relativistic focusing since the laser power used is below the critical power necessary for relativistic self-focusing at these electron densities (which is $\sim$ 2 TW at $n_e = 10^{19}$ cm^{-3}) and may be simply attributable to situations where the effects of nonlinear self-focusing [12] and ionization defocusing [13] occur simultaneously. Charge displacement or "cavitation" which was indirectly observed in a separate experiment [14] may also contribute to the self-focusing process.

It has been observed previously that the onset of ionization destroys supercontinuum structures produced by the interaction of lower intensity laser light with nonlinear media with only an ionization induced blueshift remaining in the transmitted spectrum [10]. Here, however, the target gas was a jet of hydrogen approximately 2 mm long and all of the atoms were completely ionized (the threshold for field ionization of a hydrogen atom is about 10^{14} W/cm^2). The broadening and spectral modulations of the forward scattered light may be due to the effect of self-modulation on the wavepacket by the plasma. The pulse will tend to break up into shorter pulses of about the plasma wavelength (30 fsec) and produce large amplitude plasma wakefields. If this effect becomes nonlinear - more than merely a sinusoidal modulation of the wavepacket at the electron plasma frequency - an increase in the spectral bandwidth of the forward going light may result. However, it is unclear what the source of the spectral oscillations might be and if they are related to similar oscillations observed in the stimulated Raman backscattered spectra described above. The observation of Anti-Stokes forward Raman sacttering is also indicative of large amplitude plasma wakefield generation. This has recently been reported by another group who have correlated such emission with observation of accelerated electrons [15].

It is probable that the source of the extreme broadening in the backscattered spectra is simply the high intensity limit of the SRS instability indicating that for very high intensities a much wider spectrum of unstable plasma modes can couple to the scattered light. For high intensity short pulse laser physics, parametric instabilities such as SRS are only relevant when the growth time is much less than the length of the laser pulse. Raman scattering involves the scattering of a laser photon from an electron plasma wave resulting in the transfer of energy to the plasma wave and a scattered photon. This instabilility occurs only in regions of the plasma where the electron density is less than $n_{cr}/4$. The expression for the growth rate is given by [15];

$$\gamma = \frac{a_o\,\omega_o}{4}\left[\frac{\omega_{pe}^2}{\omega_{ek}(\omega_o - \omega_{ek})}\right]^{1/2}$$

where

$$\omega_{ek} = \left(\omega_{pe}^2 + 3k^2 v_e^2\right)^{1/2}$$

is the Bohm-Gross frequency and v_e is the thermal velocity of the electrons in the plasma. For the parameters of the system at NRL this results in an e-folding time of $\approx$ 10 fsec at $n_e = 6 \times 10^{18}\ \mathrm{cm}^{-3}$, implying that for a pulse of 1 psec duration this instability should be important and may reach saturation. It should also be noted that instability thresholds depend upon the density scalelength and the damping rate of the plasma waves. This instability may also result in significant heating of electrons due to Landau damping of these waves and has been calculated to be several tens of eV for field ionized plasmas in this regime [3].

The slightly blue shifted central peak in the backscattered spectra may be the result of stimulated Brillouin scattering. Its exponential behavior with incident laser intensity can be most easily attributed to an increase in the growth rate of an instability. Stimulated Brillouin scattering is an instability in which the incident photon scatters by producing an ion acoustic wave and a lower frequency scattered photon (SBS) and can occur in plasmas up to the critical density. The expression for the growth rate is derived in Kruer [16] and is given in the strong field limit ($\omega >> kc_s$) by

$$\gamma = \frac{\sqrt{3}}{2}\left[\frac{\omega_o\, a_o^2\, \omega_{pi}^2}{2}\right]^{1/3}$$

where ω_{pi} is the ion plasma frequency. The growth rate increases with decreasing wavelength (larger k_o) and with increasing intensity (larger v_{osc}). Note that the growth rate of SBS is proportional to $I^{1/3}$,. For fully ionized hydrogen plasmas at density of 5 x 10^{18} cm^{-3} and at the maximum intensity of the NRL system this equation implies a value for γ^{-1} of 60 fsec. This instability does not cause significant heating of the plasma although it can result in considerable scattering of laser light. The fact that this light is quite intense and does not appear for intensities less than 10^{17} W/cm^2 suggest that its source is an instability and is not caused by incoherent Thomson scattering. SBS should result in a slight frequency red shift (by the ion acoustic frequency) however it has been previously observed that at higher intensity the Brillouin backscattered peak from longer pulse lasers interacting with critical density plasmas tends to blueshift [17].

It is possible that the oscillatory structure in the backscattered spectra is due to ion fluctuations in the plasma. Note that the ion plasma period for hydrogen is 1.8 psec at these densities while that for helium is 2.5 psec, and they are close to the oscillation periods depicted in Fig. 4. While the oscillation frequency seemed to correlate to the ion plasma period approximately, there remains uncertainty in these measurements primarily due to a 25% error in the density measurements. However it is apparent that several fundamental frequencies exist and that there are clear differences between the Fourier spectrum of hydrogen and helium at similar electron densities. In most plasmas, ion plasma waves are severely damped - however if large electron plasma wakefields are simultaneously created in the medium the backscattered SRS light may travel through regions of great plasma turbulence where a large spectrum of waves have been excited - including ion plasma oscillations which may modulate the the spectrum of the backscattered light.

In conclusion, these experiments have resulted in new observations of high intensity laser scattering mechanisms in field ionized plasmas. Forward continuum generation was measured in an ionized medium which may be due to self-modulation effects of the wavepacket by the plasma. Stimulated - or coherent instabilities such as SRS and SBS seem to occur at a much greater efficiency than Thomson scattering and hence are the source of most of the scattered signal - although Brillouin scattering is somewhat weaker than Raman scattering. The high intensity limit of the SRS instability results in the generation of an extremely broad, supercontinuum like structure in the backscattered direction. It appears that significant ion plasma fluctuations can be excited in such plasmas which can modulate the spectrum of the backscattered Raman emission.

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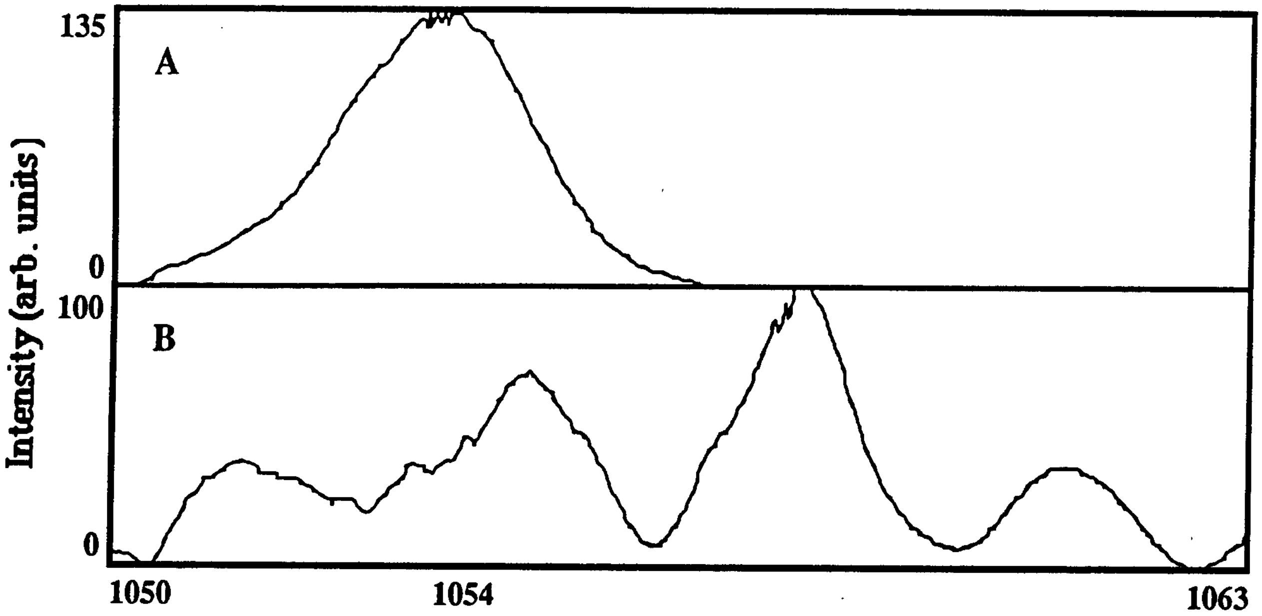

FIG. 1 Broadening and oscillatory structure of forward scattered laser light near the fundamental, A) original spectrum of laser pulse (~3 nm in width), B) spectrum of transmitted light.

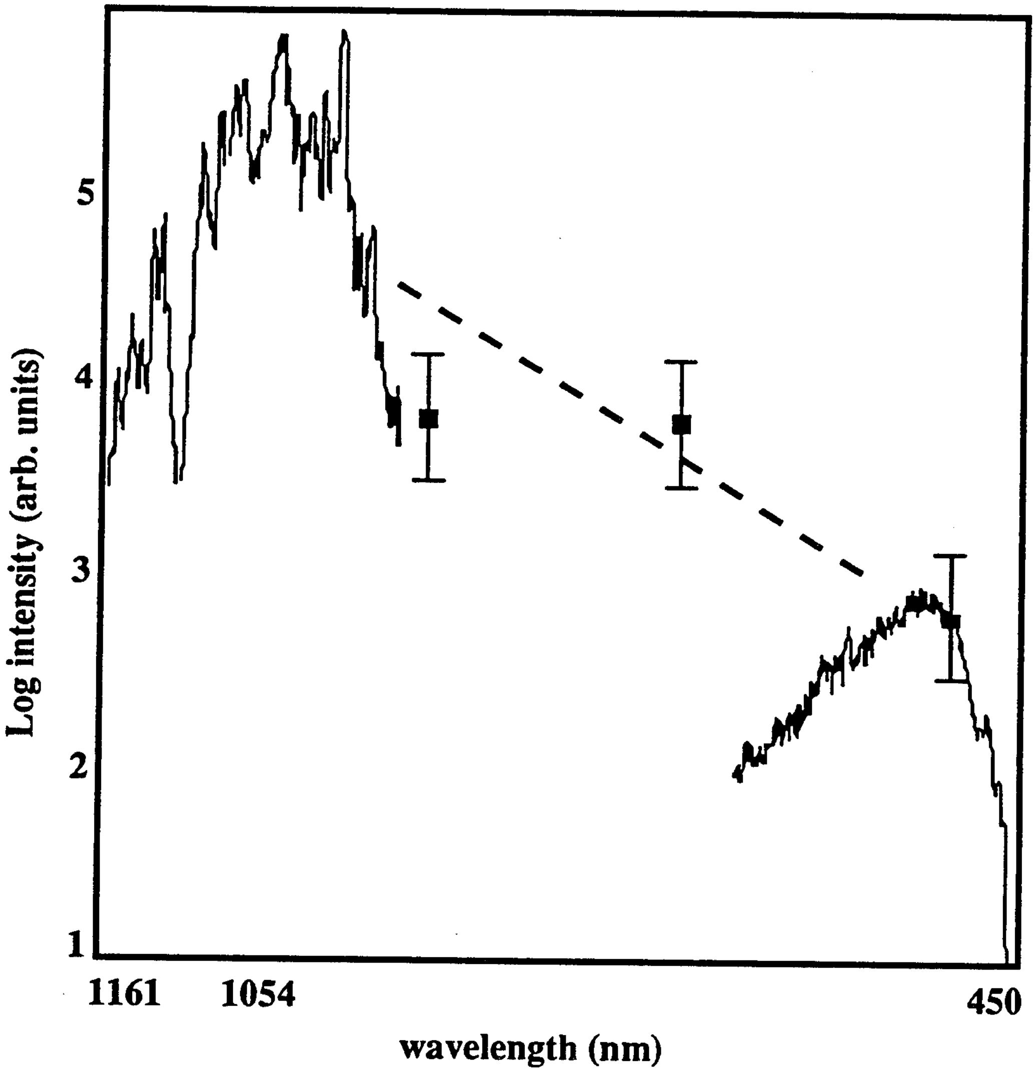

FIG. 2 Broad Raman backscattered spectrum (composite of several spectra, data points are condensed narrow bandwidth spectra) - note that the apparent decrease in the spectrum in the region near 600 nm is due to the combined effects of filters, diffraction grating efficiency and detector efficiency in this region.

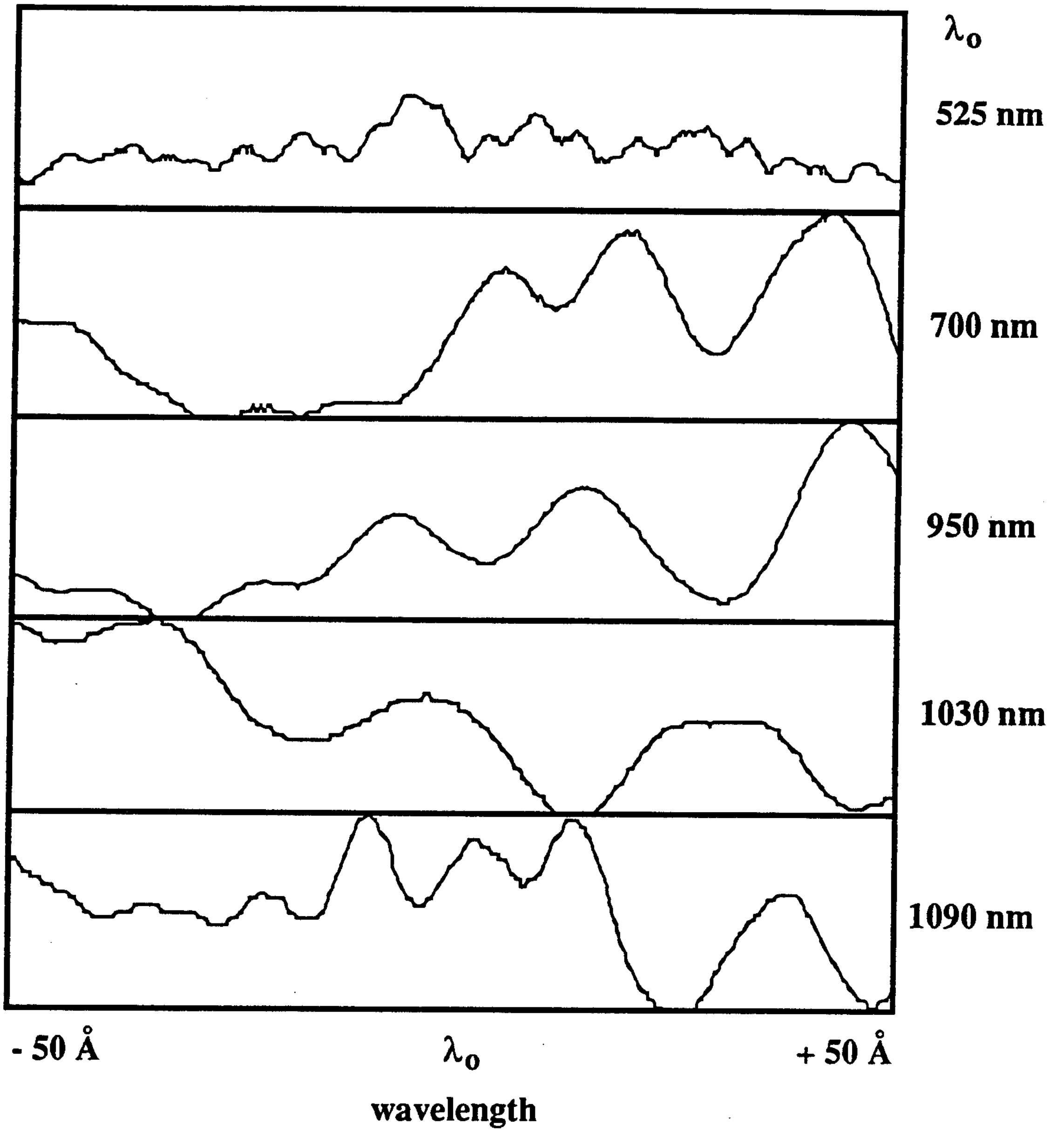

FIG. 3 Behavior of directly backscattered spectrum across frequency range. The bottom two spectra were taken with OD 4 filters.

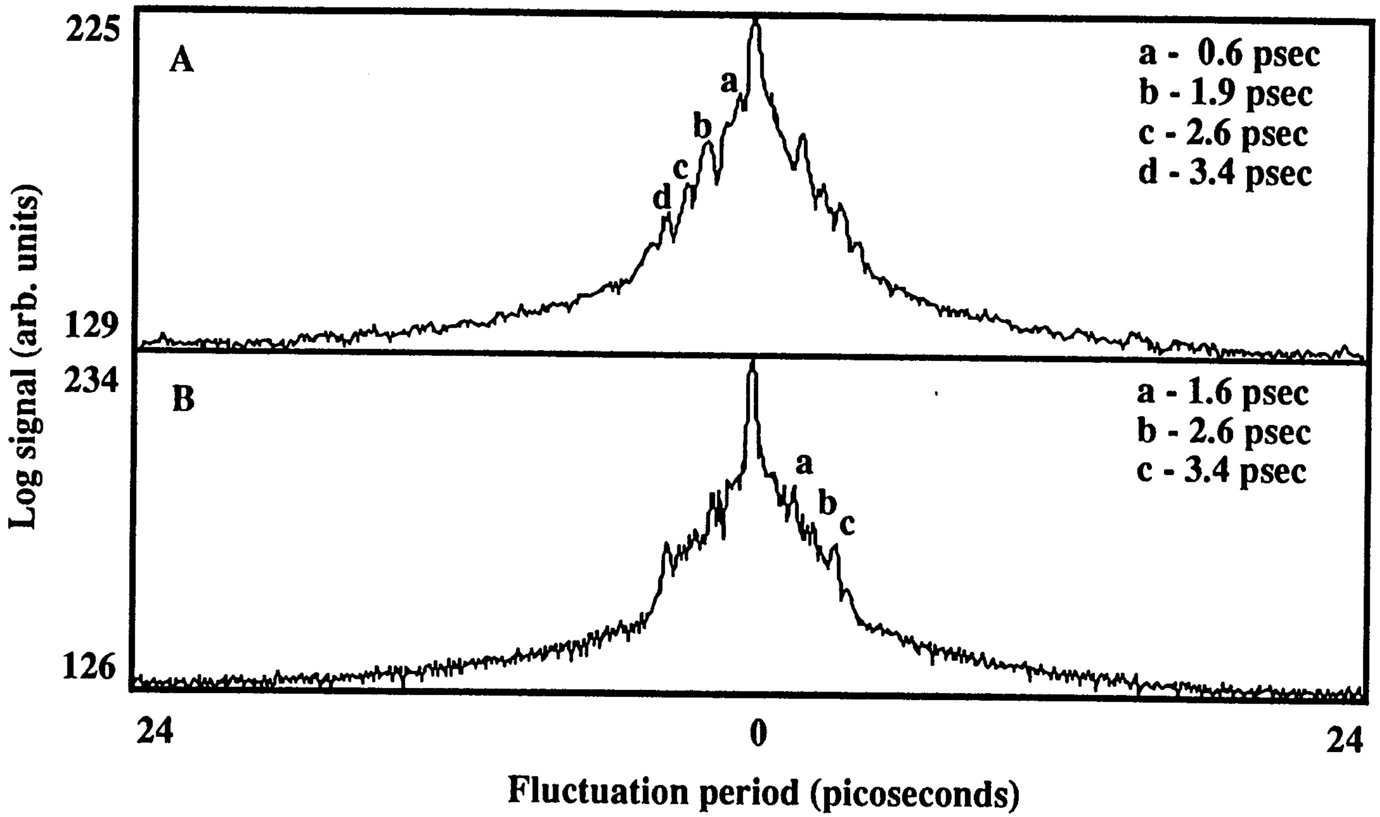

FIG. 4 Fourier transform of backscattered frequency spectrum: A) hydrogen, B) helium.