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M.-A. Mycek, U. Siegner, and D.S. Chemla

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INSTANTANEOUS FREQUENCY DYNAMICS OF COHERENT EMISSION FROM MAGNETOEXCITONS IN GALLIUM ARSENIDE

Mary-Ann Mycek[1,2], Uwe Siegner[2], and Daniel S. Chemla[1,2]

[1]Department of Physics
University of California
Berkeley, CA 94720

[2]Materials Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

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Mary-Ann Mycek, Uwe Siegner, Daniel S. Chemla

Department of Physics, University of California at Berkeley,

and

Materials Sciences Division, Lawrence Berkeley Laboratory, University of California

Berkeley, California 94702

Phone: (510)-486-5265 *FAX: (510)-486-5530*

Abstract:

By time resolving the power spectrum of the ultrafast four wave mixing emission from the lowest magnetoexciton in GaAs, we determine the origin of the very complicated dynamics observed in the time domain.

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Department of Physics, University of California at Berkeley,

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In semiconductors, the application of a magnetic field B dramatically influences the linear and nonlinear optical properties. Our measurements on GaAs indicate that the transition from a 3D to a 1D system under field provides the ideal laboratory for studying both the dephasing properties of and interactions between magnetoexcitons.

The linear absorption spectrum of a high quality sample is shown in Fig.1 for (a) B = 0 and (b) B = 6T. Strain has split the heavy hole (HH) and light hole (LH) exciton, with the HH resonance at higher energy [1]. At B=6T, the coupling between higher order magnetoexciton states and the 1D continua produces

pronounced Fano resonances. [2] Here, we concentrate on the dynamics of the lowest magnetoexciton. Experiments were performed at 1.6K and B = 6T using 100fs laser pulses (dashed curve, Fig.1).

The ultrafast nonlinear optical magnetoexciton response was measured by *spectrally and temporally* resolving the coherent four wave mixing (FWM) emission. The temporal response is presented in Fig.2, which shows the time-integrated (TI) FWM signal measured at three excitation densities. For all densities, the TI-signal has a contribution at negative time delays (Δt) and a non-exponential decay for positive Δt. These features are density dependent and are particularly evident in Fig.2c at the lowest density $N \approx 10^{16}$ cm^{-3}. Here, the dephasing time becomes very long (the TI-signal extends beyond 3ps) and a complicated beating structure is apparent for positive and negative Δt.

This unusual response can be better understood by monitoring the spectrum of the FWM emission. In fact, the presence of each beat node in the temporal profile can be traced to a spectral change. This is shown in Fig.3, where Fig.3a is the natural logarithm of the TI-FWM of Fig.2c. For Δt around the beat node at $\approx$ -300fs (dashed box), we present the spectra of the FWM emission in Figs.3b-3f, plotted on the same scale. Before the beat node, Figs.3b,3c, the emission is dominated by the HH-exciton at λ=818nm, which is flanked by smaller contributions. Fig.3d, close to the beat node, shows the total emission significantly reduced, with three small peaks having nearly the same intensity. After the beat node, Fig.3e,3f, the total emission increases rapidly, but now the spectrum is dominated by a low energy contribution lying between the LH and HH exciton. Thus, the origin of the beating is a quantum interference between spectral components. Such spectral characteristics are known to affect not only the

amplitude, but also the phase of the FWM emission. [3] In fact, here we observe the extremely fast rate of change in the FWM instantaneous frequency reflected directly on the temporal profile as the weight of the spectral contributions changes.

Importantly, one may use the spectral data to measure the intensity of a particular component vs. time delay and extract magnetoexciton dephasing times even though the temporal profile is too complicated to directly analyse. In this way, the HH exciton was determined to have a decay time $\geq$ 1.1 ps. In contrast, the low energy contribution decays much faster, in $\approx$ 90fs. These density dependent ultrafast dynamics reveal the complexity of magnetoexciton interactions in GaAs. Results for B = 0 - 12 Tesla will also be presented.

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Figure Captions:

Figure 1: Linear absorption spectra for our GaAs sample at 1.6K taken with circularly polarized light: (a) B = 0, (b) B = 6 Tesla. The spectrum of the excitation pulse used in the FWM experiments is shown in the dashed line.

Figure 2: Time integrated FWM measurements performed in the two-pulse geometry using co-circularly polarized light for excitation densities: (a) $N \approx 5 \times 10^{16}$ cm^{-3}, (b) N / 1.6, (c) N / 5.

Figure 3: For excitation density $N \approx 10^{16}$ cm^{-3}, (a) shows the time integrated FWM measurement, while (b)-(f) present the corresponding FWM power spectra for time delays:
(b) -560fs, (c) -400fs, (d) -320fs, (e) -240fs, (f) -160fs.

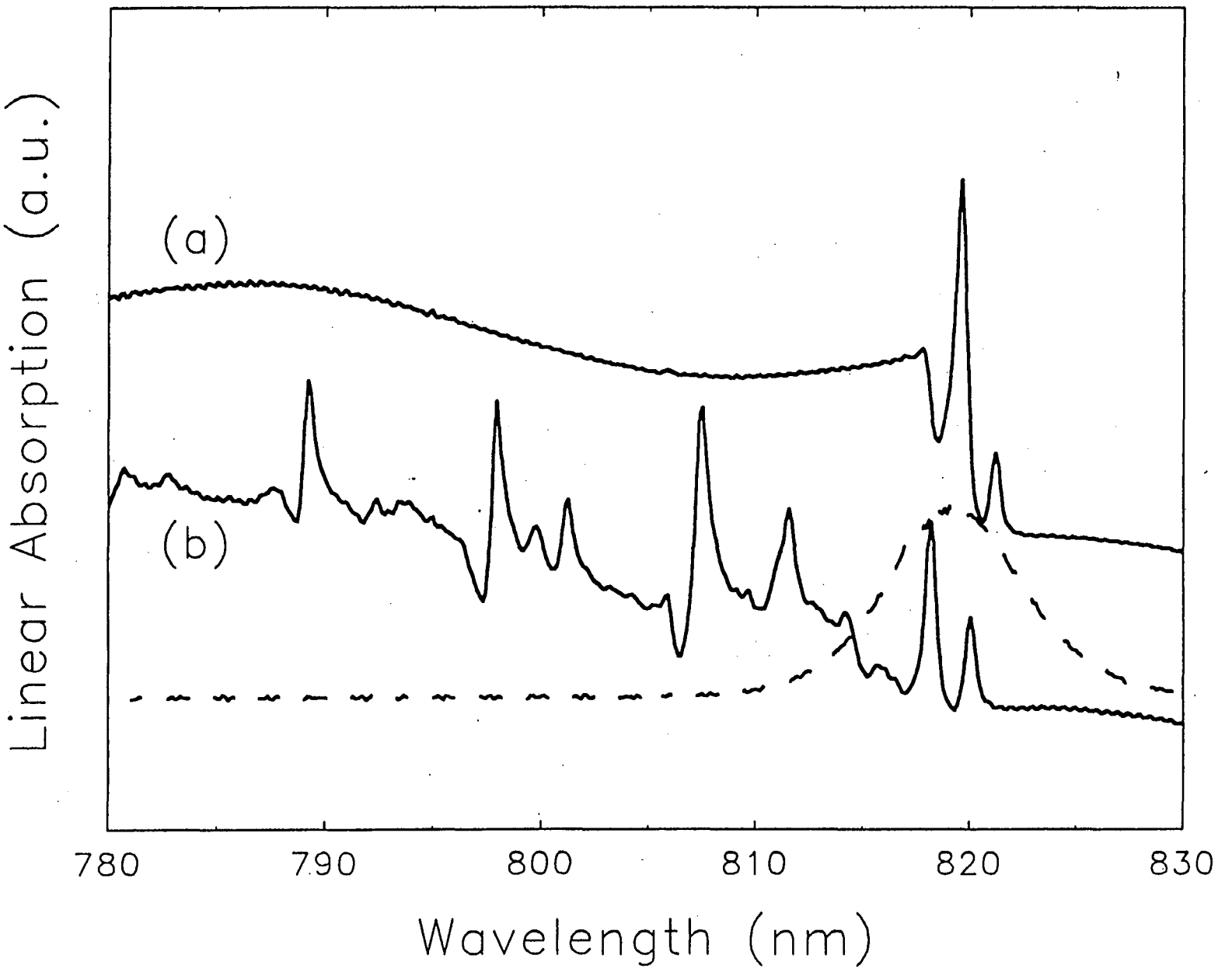

FIG. 1

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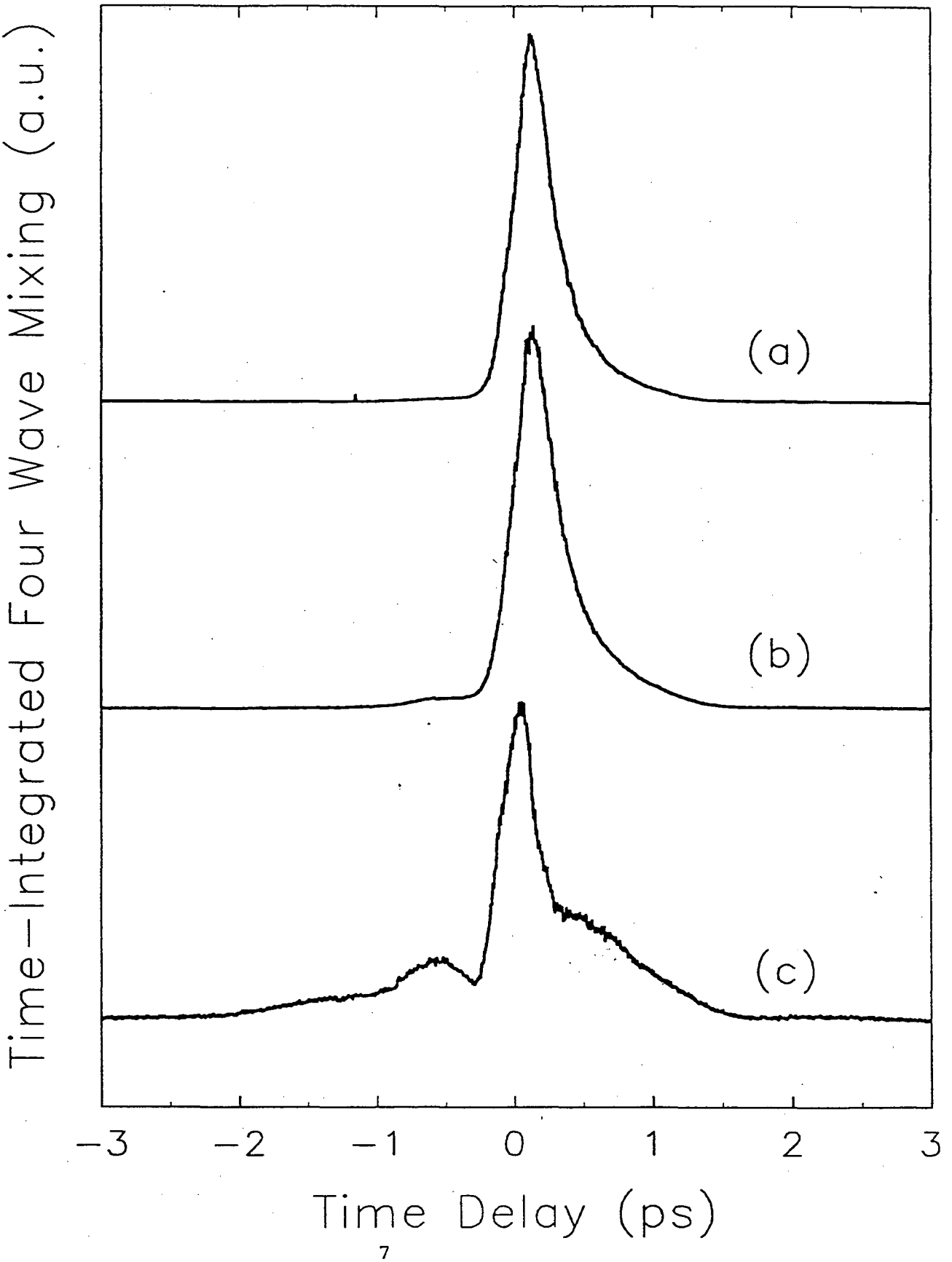
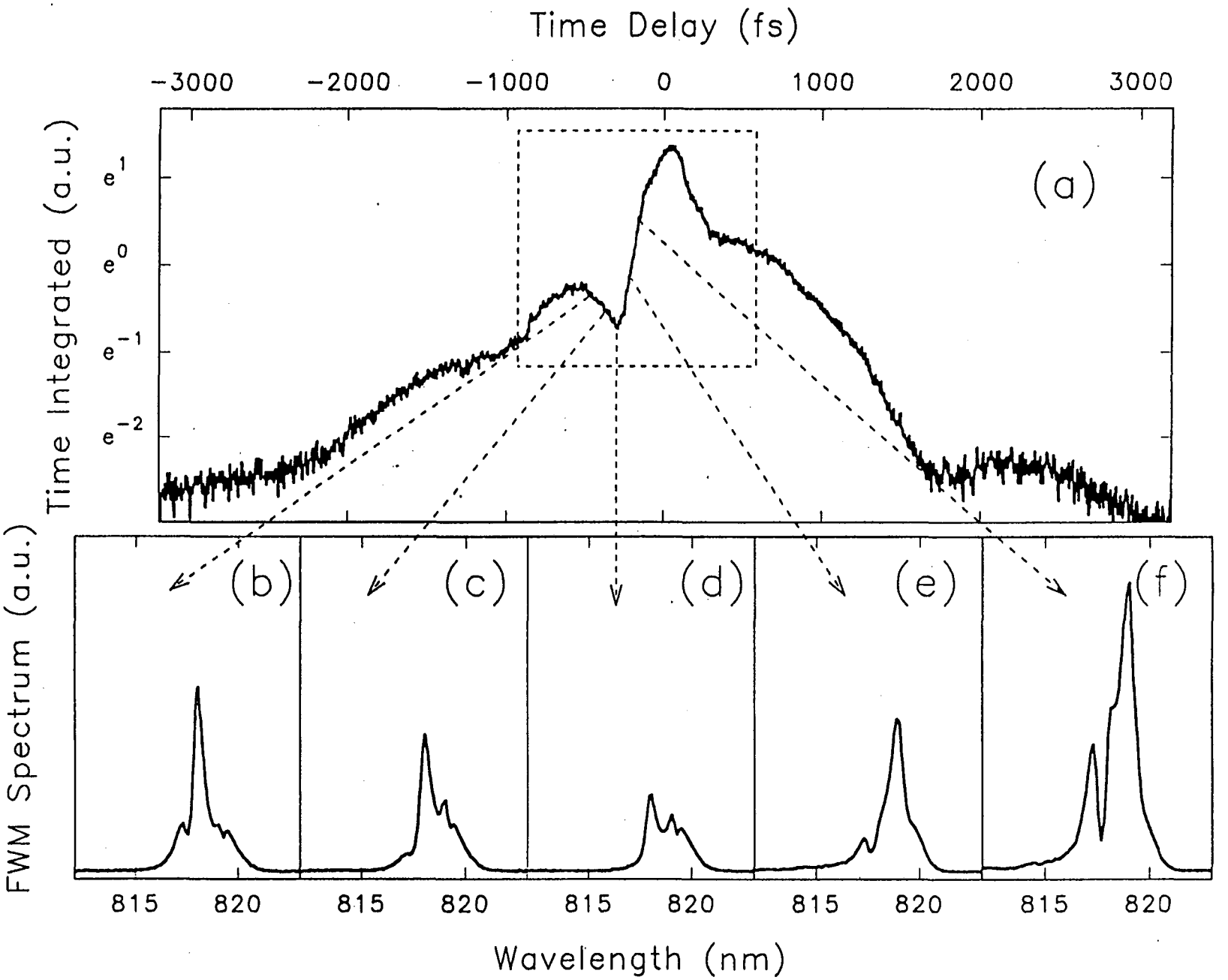

Time-Integrated Four Wave Mixing (a.u.)
(a)
(b)
(c)
−3
−2
−1
0
1
2
3
Time Delay (ps)


Time Delay (fs)
−3000
−2000
−1000
0
1000
2000
3000
Time Integrated (a.u.)
e^1
e^0
e^−1
e^−2
(a)
(b)
(c)
(d)
(e)
(f)
FWM Spectrum (a.u.)
815
820
Wavelength (nm)

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
TECHNICAL INFORMATION DEPARTMENT
BERKELEY, CALIFORNIA 94720