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## Figure Captions for Supplementary Material

**Figure S1.** Transient resonance Raman spectra of the ethynyl region of compound **I** in THF obtained with pulsed excitation at 436 nm. (A) result of subtraction of spectrum (C) from spectrum (B); scaling factor = 8.26; (B) high power (28 mW) spectrum; (C) low power (2.1 mW) spectrum. Excitation wavelength = 436 nm, pulse width = 5 ns, T = 25 °C.

**Figure S2.** Transient resonance Raman spectra of the ethynyl region of compound **I** in THF obtained with pulsed excitation at 436 nm. (A) result of subtraction of spectrum (C) from spectrum (B); scaling factor = 7.08; (B) high power (28 mW) spectrum; (C) low power (2.1 mW) spectrum. Excitation wavelength = 436 nm, pulse width = 5 ns, T = 25 °C. Digital subtraction of the spectra was performed with LabCalc version 2.0 (Galactic Industries). Prior to subtraction, the baseline of each spectrum was corrected with the "multiple point" routine in LabCalc. A scaling factor was chosen so that no negative peaks appeared in the difference spectrum. The second-derivative-like feature in the difference spectrum is weak but reproducible, thus indicating broadening of the wings of the ethynyl mode upon high-intensity laser excitation.

**Figure S3.** Transient resonance Raman spectra of the ethynyl region of compound **II** in THF obtained with pulsed excitation at 436 nm. (A) result of subtraction of spectrum (C) from spectrum (B); scaling factor = 2.47; (B) high power (16 mW) spectrum; (C) low power (1.2 mW) spectrum. Excitation wavelength = 436 nm, pulse width = 5 ns, T = 25 °C.

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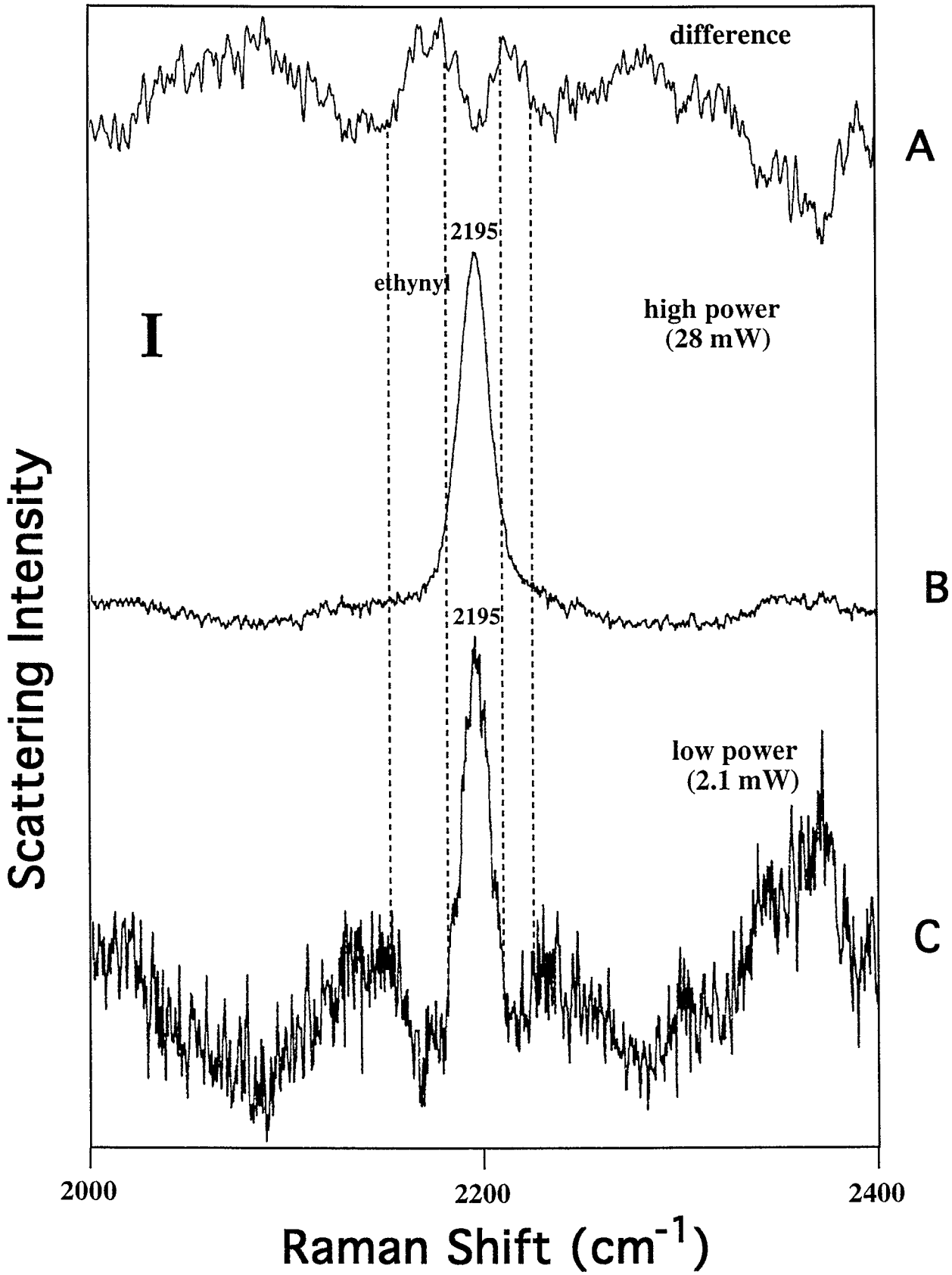
**Figure S4.** Transient resonance Raman spectra of the ethynyl region of compound **II** in THF obtained with pulsed excitation at 436 nm. (A) result of subtraction of spectrum (C) from spectrum (B); scaling factor = 2.55; (B) high power (16 mW) spectrum; (C) low power (1.2 mW) spectrum. Excitation wavelength = 436 nm, pulse width = 5 ns, T = 25 °C. Digital subtraction of the spectra was performed with LabCalc version 2.0 (Galactic Industries). Prior to subtraction, the baseline of each spectrum was corrected with the "multiple point" routine in LabCalc. A scaling factor was chosen so that no negative peaks appeared in the difference spectrum. The second-derivative-like feature in the difference spectrum is weak but reproducible, thus indicating broadening of the wings of the ethynyl mode upon high-intensity laser excitation.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A
2195
ethynyl
I
high power
(28 mW)
B
2195
low power
(2.1 mW)
C
Scattering Intensity
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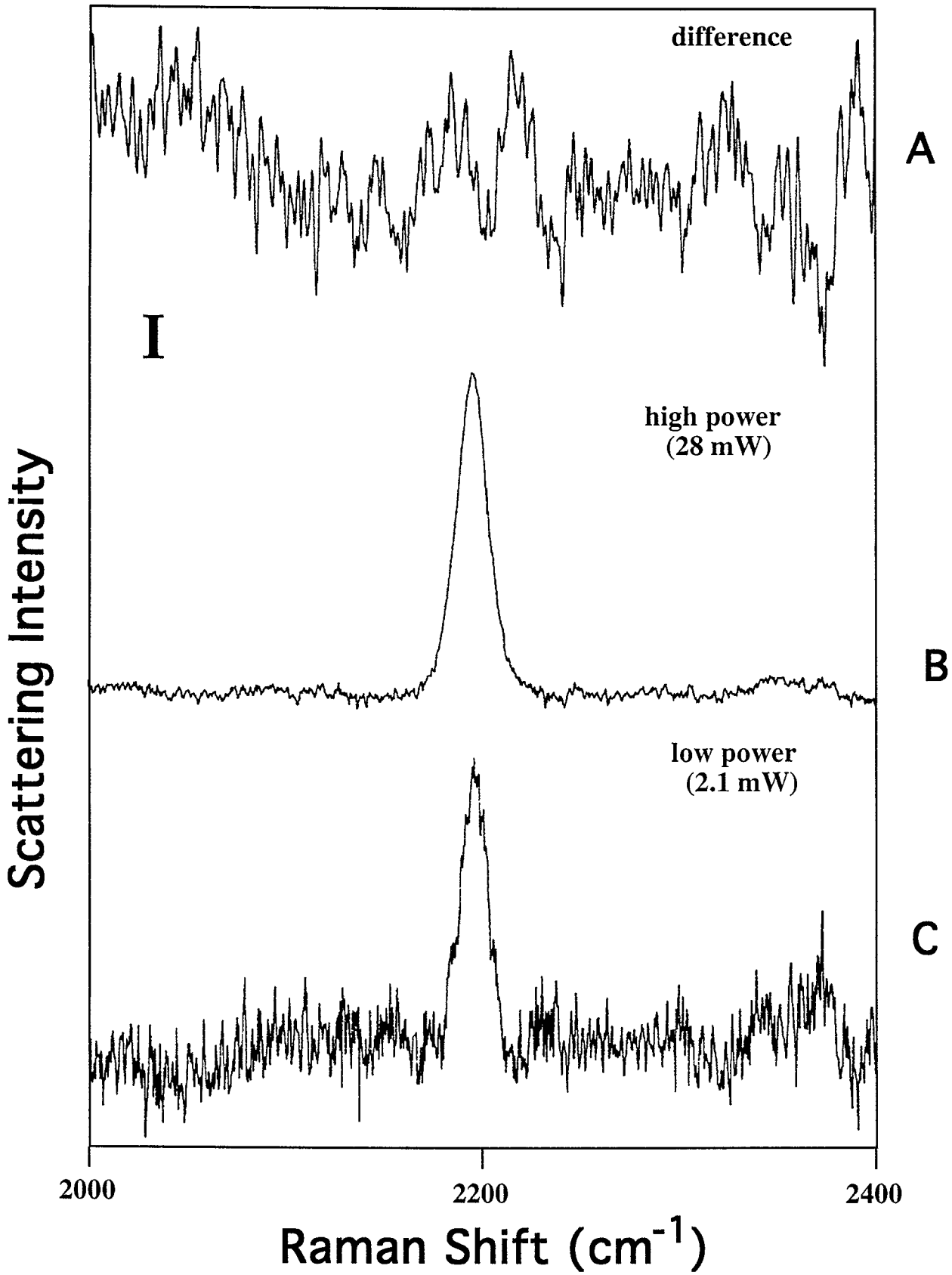
2200
2400
Raman Shift (cm-1)


difference
A
I
high power
(28 mW)
B
low power
(2.1 mW)
C
Scattering Intensity
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2200
2400
Raman Shift (cm-1)

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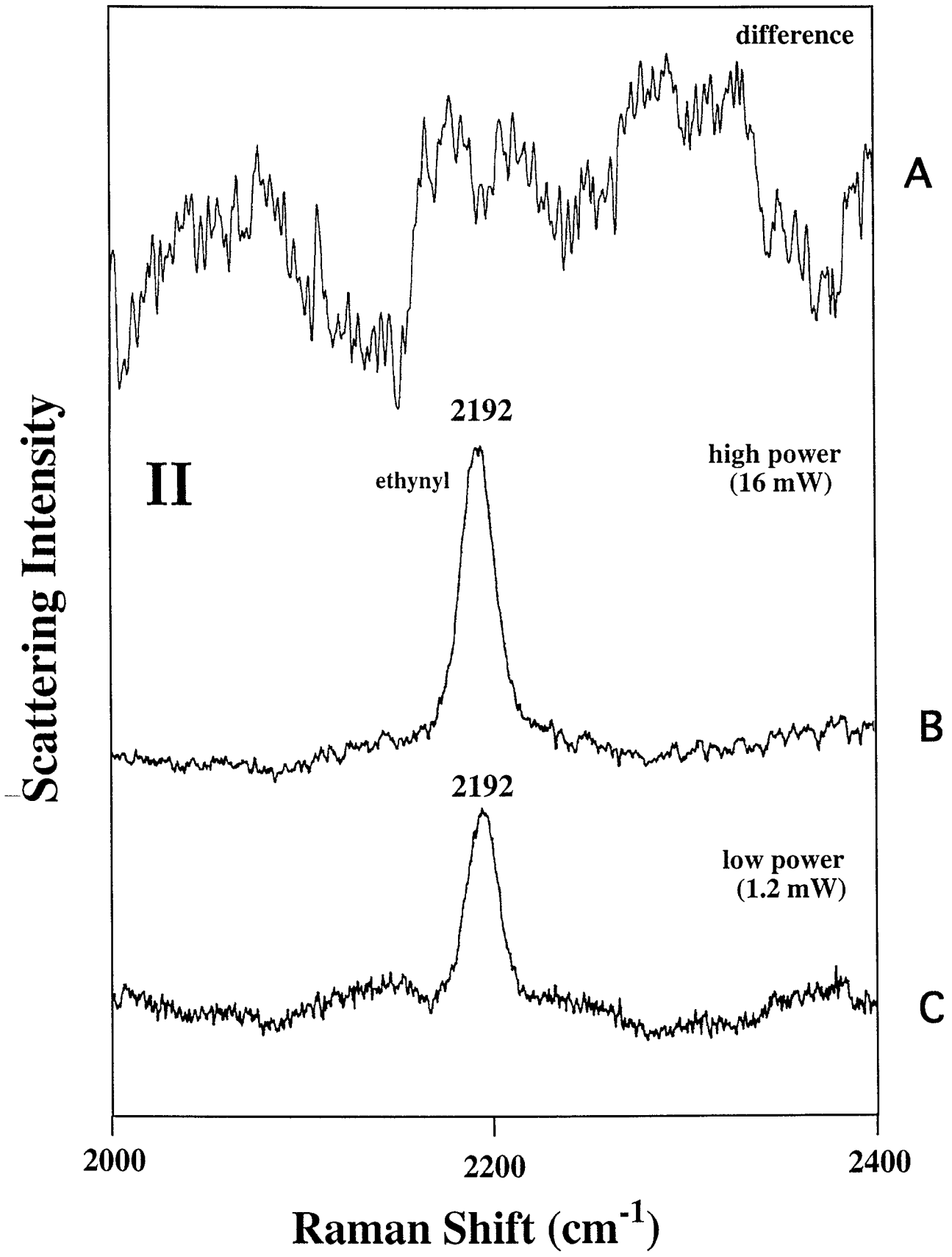

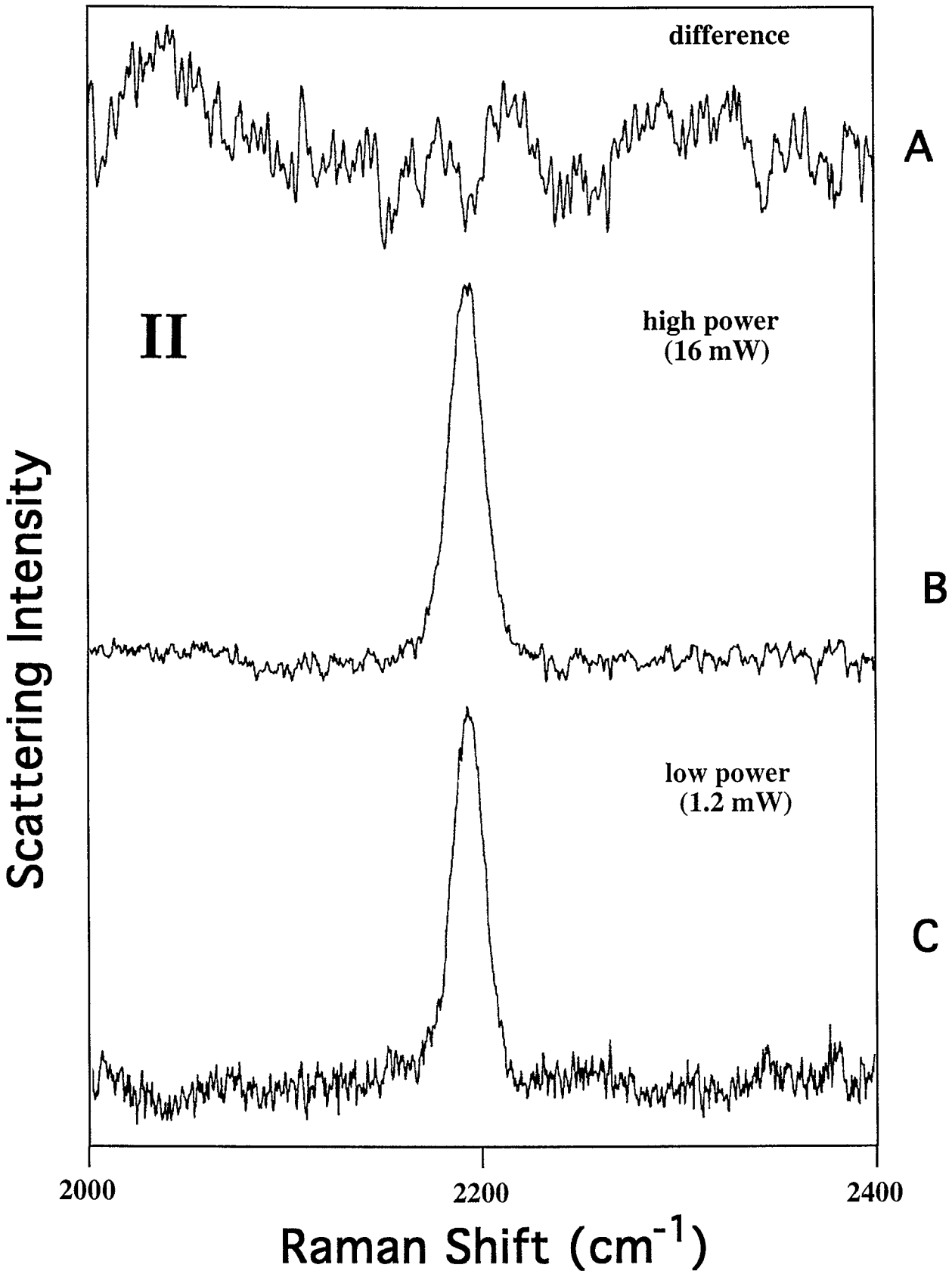
difference
A
II
high power
(16 mW)
B
low power
(1.2 mW)
C
Scattering Intensity
2000
2200
2400
Raman Shift (cm-1)