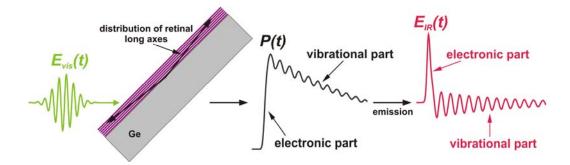
## Vibrational motions associated with primary processes in bacteriorhodopsin studied by coherent infrared emission spectroscopy

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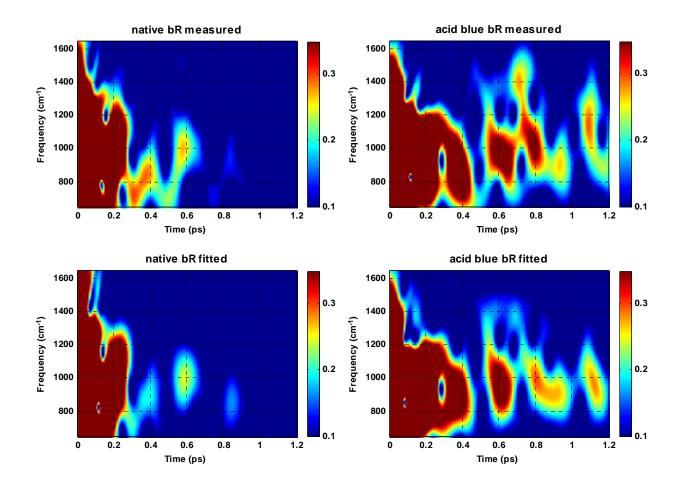
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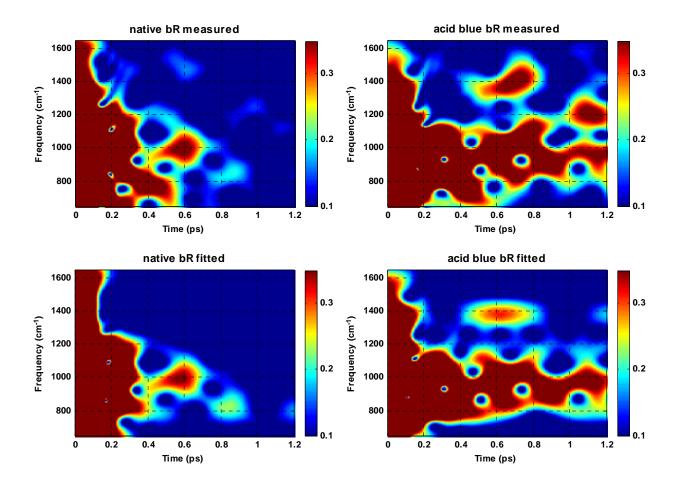
## **Supplemental figures**



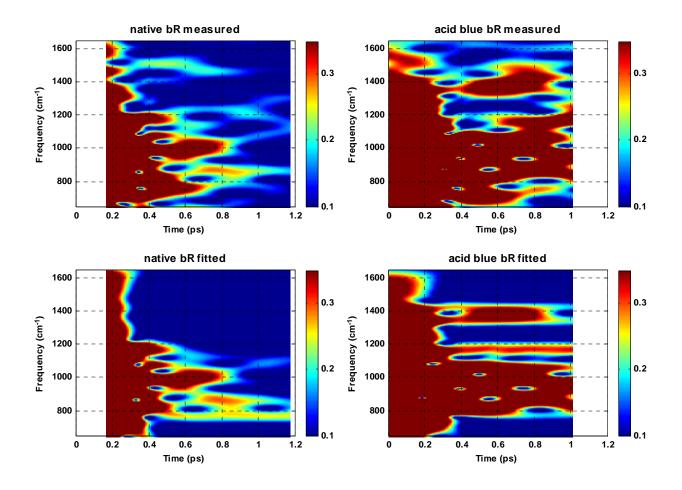
*Figure S1.* Principle of the generation of coherent infrared emission from oriented bacteriorhodopsin films. As a response to excitation with an ultrashort visible laser pulse, a polarization is created in the non-centrosymmetric sample. This time-dependent polarization P(t) generates an electromagnetic wave with electric field  $E_{IR}(t)$  proportional to its first derivative. This wave is composed of an electronic part originating from the instantaneous electronic polarization, and a long-lasting vibrational response. The emitted beam is polarized with a component in the direction of the symmetry axis of the film, and there is no intensity in this direction. Therefore, the sample is tilted with respect to the plane perpendicular to the beams.



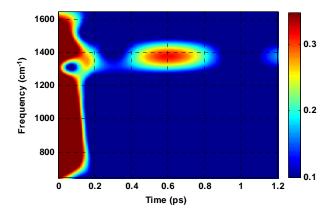
*Figure S2.* Spectrogram of the interferogram of the native the acid blue bR sample with GaAs reference, based on sliding window Fourier analysis with Hann window of 200 fs length (100 fs FWHM).



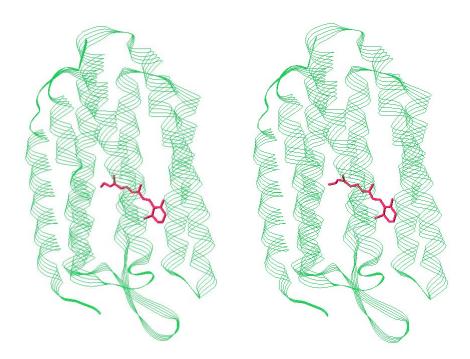
*Figure S3.* Spectrogram of the interferogram of the native the acid blue bR sample with GaAs reference, based on sliding window Fourier analysis with Hann window of 400 fs length (200 fs FWHM).



*Figure S4.* Spectrogram of the interferogram of the native the acid blue bR sample with GaAs reference, based on sliding window Fourier analysis with Hann window of 800 fs length (400 fs FWHM).



*Figure S5.* Effect of beating of two modes on the spectrogram (Hann window of 400 fs length): acid blue bR reconstructed from fitting, taking into account only the electronic process and the two modes of the highest frequency ( $1361 \text{ cm}^{-1}$  and  $1408 \text{ cm}^{-1}$ ).



*Figure S6.* Arrangement of the retinal chromophore in the bR molecule. Left: resting state of native bR with *all-trans* retinal isomer (Protein Database entry code 1IW6). Right: Long-lived K intermediate with *13-cis* retinal isomer (Protein Database entry code 1IXF). In both cases the retinal structure is highly planar and perpendicular to the horizontal plane of the membrane.