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Title:Pulsed electron paramagnetic resonance spectroscopy powered by a free-electron laser

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Abstract:Electron paramagnetic resonance (EPR) spectroscopy interrogates unpaired electron spins in solids and liquids to reveal local structure and dynamics; for example, EPR has elucidated parts of the structure of protein complexes that other techniques in structural biology have not been able to reveal. EPR can also probe the interplay of light and electricity in organic solar cells and light-emitting diodes, and the origin of decoherence in condensed matter, which is of fundamental importance to the development of quantum information processors. Like nuclear magnetic resonance, EPR spectroscopy becomes more powerful at high magnetic fields and frequencies, and with excitation by coherent pulses rather than continuous waves. However, the difficulty of generating sequences of powerful pulses at frequencies above 100 gigahertz has, until now, confined high-power pulsed EPR to magnetic fields of 3.5 teslas and below. Here we demonstrate that one-kilowatt pulses from a free-electron laser can power a pulsed EPR spectrometer at 240 gigahertz (8.5 teslas), providing transformative enhancements over the alternative, a state-of-the-art ~30-milliwatt solid-state source. Our spectrometer can rotate spin-1/2 electrons through $\pi/2$ in only 6 nanoseconds (compared to 300 nanoseconds with the solid-state source). Fourier-transform EPR on nitrogen impurities in diamond demonstrates excitation and detection of EPR lines separated by about 200 megahertz. We measured decoherence times as short as 63 nanoseconds, in a frozen solution of nitroxide free-radicals at temperatures as high as 190 kelvin. Both free-electron lasers and the quasi-optical technology developed for the spectrometer are scalable to frequencies well in excess of one terahertz, opening the way to high-power pulsed EPR spectroscopy up to the highest static magnetic fields currently available.

Number of references:30

Inspec controlled terms:diamond - EPR spectrometers - EPR spectroscopy - Fourier transform

spectroscopy - free electron lasers - free radicals - impurities - magnetic fields - nitrogen
Uncontrolled terms:pulsed electron paramagnetic resonance spectroscopy - free-electron laser -
confined high-power pulsed EPR spectroscopy - one-kilowatt pulses - pulsed EPR spectrometer -
transformative enhancements - spin-1/2 electrons - Fourier-transform EPR - nitrogen impurities -
decoherence times - frozen solution - nitroxide free-radicals - quasioptical technology - terahertz
frequencies - gigahertz frequencies - static magnetic fields - frequency 240 GHz - magnetic flux
density 8.5 tesla - C:N

Inspec classification codes:A0758 Magnetic resonance spectrometers, auxiliary instruments and
techniques - A4255T Free electron lasers - A0765 Optical spectroscopy and spectrometers -
A6170W Impurity concentration, distribution, and gradients - A7630L EPR of other ions and
impurities - A7630R EPR of free radicals (condensed matter)

Numerical data indexing:frequency 2.4E+11 Hz;magnetic flux density 8.5E+00 T

Chemical indexing:C:N/bin C/bin N/bin C/el N/el N/dop

Treatment:Practical (PRA); Experimental (EXP)

Discipline:Physics (A)

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