SCANNING PROBE MICROSCOPY

Close-up on spin coherence

Ultrafast, coherent spin dynamics in semiconductor heterostructures can be measured with a scanning tunnelling microscope by using femtosecond pulses of circularly polarized light.

Sebastian Loth, Jacob A. J. Burgess and Shichao Yan

Spins are the basic buildings blocks of magnetism, and exploring their dynamics and interactions is central to understanding technologically relevant materials ranging from magnetic nanodots to high-temperature superconductors. The elementary processes that govern the dynamics of spins typically occur on length scales of ångströms to nanometres and with characteristic timescales of femto- to nanoseconds. So far, this combination of time and spatial resolution has been an experimental blind spot. Far-field techniques offer sufficient time resolution[1,2] but, despite intensive effort, have not yet reached a resolution in real space better than 10 nm. Scanning probe techniques, on the other hand, readily resolve magnetic structures with subnanometre spatial resolution, but are typically limited to slow, millisecond speed and their use restricted to investigations of quasi-static surface magnetism. Writing in *Nature Nanotechnology*, Hidemi Shigekawa and colleagues at Tsukuba University now show that ultrafast coherent magnetization dynamics are within the reach of the scanning tunnelling microscope[3].

Recently, the time resolution of scanning tunnelling microscopy (STM) has been significantly improved by implementing pump–probe measurement schemes to measure the lifetime of photoexcited carriers[4,5] and nanosecond spin relaxation in few-atom nanomagnets[6]. Shigekawa and co-workers interface ultrafast optics in a pump–probe scheme with cross-sectional STM to resolve coherent spin dynamics on the picosecond timescale within a quantum well only 6 nm in width (Fig. 1). They measured the spin relaxation of electrons in GaAs/AlGaAs quantum wells on a picosecond timescale and the quantum beat of the electron spin in a magnetic field. This marks the first time that coherent spin dynamics have been captured with a scanning tunnelling microscope, adding laser-combined STM to the list of ultrafast magnetization probes.

The researchers cleaved a piece of a GaAs-based heterostructure wafer in vacuum and approached the freshly exposed surface with the microscope's tip. Buried interfaces and heterostructures can be accessed in this geometry and the team exploited this capability to make measurements across individual quantum wells. Ultrafast pulses of circularly polarized light from Ti:sapphire lasers were focused onto the sample around the tip causing electron–hole pairs to be photoexcited. The holes quickly relaxed and were not measured. The hole relaxation leaves an accumulation of electrons that have a net spin polarization, due to the selection rules for photoexcitation with circularly polarized light. The spins lose their orientation by scattering with impurities or other free carriers over a characteristic time called the spin-relaxation time.

To detect this electron-spin relaxation time the researchers make use of saturation effects in the excitation of free carriers to the conduction band. An initial laser pulse (the pump pulse) creates spin-polarized electrons. The second laser pulse (the probe pulse) does the same, but with an efficiency that depends on the persistence of the population of spin-polarized electrons from the pump pulse. If the probe pulse occurs at the same time as the pump pulse, or before the spin-polarized electrons have fully relaxed, fewer electrons will be excited. The tunnel current cannot be measured at the speed of the optical pulses, but depends on the average excitation efficiency of the probe pulse. Hence, the ultrafast spin relaxation can be recorded by monitoring the change of

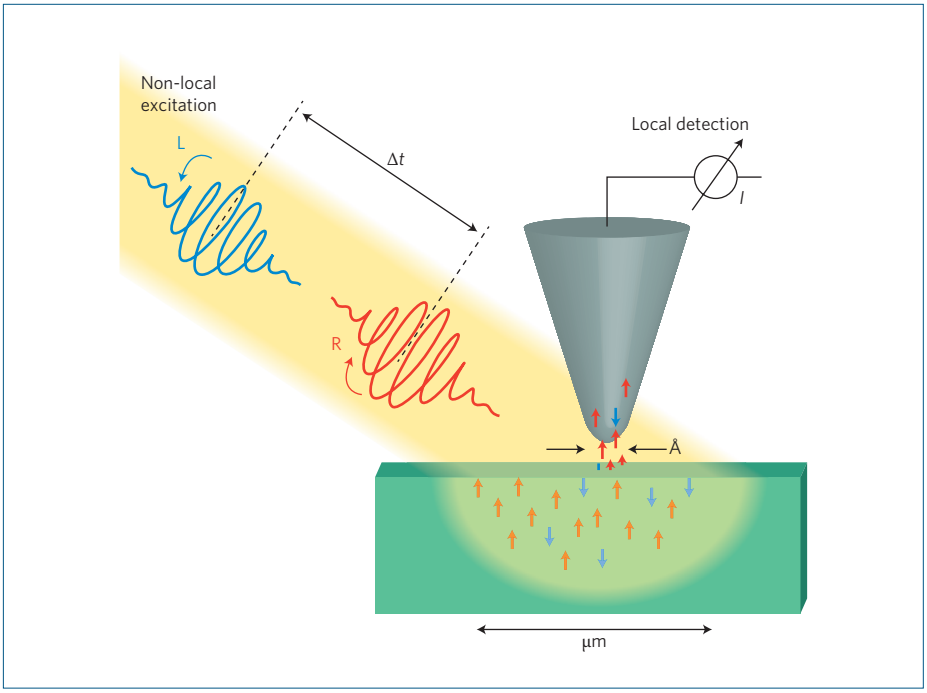

Figure 1 | Spin-sensitive pump–probe spectroscopy in the scanning tunnelling microscope. Ultrafast laser pulses with right-circular (R) and left-circular (L) polarization excite spin-polarized carriers close to the surface of a GaAs sample. The tip of a scanning tunnelling microscope is positioned in the micrometre-sized laser focus and locally measures the population of photogenerated carriers with significantly higher spatial resolution of a few ångströms. Because the spin polarization (imbalance of up and down spins) diminishes with the characteristic spin-relaxation time, the tunnel current magnitude, *I*, depends on the time delay between the pump and probe pulses, Δ*t*.

the tunnel current while slowly varying the time delay between pump and probe.

As with many experiments using near-field probes, the key to success lies in suppressing spurious effects while boosting the desired signal above the noise floor. Shigekawa and colleagues demonstrate a clever lock-in detection scheme akin to heterodyne detection in a radio receiver to boost their sensitivity and isolate the spin-dependent signal while keeping the average laser power impinging on the tip constant. Maintaining a constant power that is absorbed by the tip is an essential step to avoid spurious signals related to time-dependent heating, which have plagued laser-combined STM in the past[7].

An important aspect of the experiment is the non-local nature of the optical excitation. The micrometre-sized laser beam presents a global excitation compared with the tunnel current detection, which is localized on the ångström scale. It is possible that many photoexcited electrons, not just those generated immediately under the scanning tunnelling microscope tip, are detected at each pump–probe cycle. Consequently, this measurement scheme may also be applicable to the study of non-local effects, such as spin diffusion in real space. It also opens the door to take other established methods in ultrafast optics to the nanoscale, such as the resonant excitation of phonons[8]. Reliance on a global excitation, however, also creates a challenge for other sample systems, such as metals, where vanishing optical penetration depth significantly reduces the excitation volume and may preclude the application of laser-combined STM. Surmounting this challenge will be important for expanding the scope of this technique. Local enhancement of the light intensity under the tip, as achieved, for example, with plasmonic waveguides[9], could overcome limitations in excitation efficiency and reach the same highly localized control as approaches employing fast gating of the tunnel junction voltage[5,6].

Perhaps the most important finding of the work by Shigekawa and colleagues is the measurement of spin dynamics beyond population decay, showing that the scanning tunnelling microscope can detect coherent spin precession with high fidelity. Hallmark features are the detection of the quantum beat of the spin polarization in a magnetic field[10] and its resonant amplification with matched laser-pulse repetition[11].

Electron spins in semiconductors can maintain quantum–mechanical coherence only for short periods of time. In this time, they can, in principle, be used for spintronic applications that go further than static magnetoresistance. The method demonstrated by Shigekawa and colleagues is the first that is sensitive to spin coherence at nanometre length-scales. Decoherence and relaxation of spins in semiconductors is governed by nanometre-sized interactions with defects and disorder potentials. Understanding these processes is of key importance in the engineering of devices that harness spin coherence exceeding nanoseconds. With the local detection mechanism established, many questions could now be answered. For example, how spatially uniform is the spin coherence? What role do defects play in the emission of spin waves? What is the influence of interface disorder on spin injection across device layers?

Spatial heterogeneity in solids is intricately linked to the observable dynamics therein. Combining STM with pump–probe measurement schemes, as exemplified by the work of Shigekawa and colleagues, presents a new generation of scanning probe experiments that access elementary processes at their intrinsic length- and timescales. ❐

Sebastian Loth, Jacob A. J. Burgess and Shichao Yan are at the Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany, and the Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany.
e-mail: sebastian.loth@mpsd.mpg.de

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GRAPHENE

Electrons en masse

Massless electrons in graphene exhibit a mass when considered as collective excitations known as plasmons.

Fengnian Xia

A plasmon is the quantum of collective motion of charged carriers and typical plasmonic materials are noble metals such as silver and gold, which have many free electrons[1,2]. Plasmons have attracted significant interest recently due to their ability to confine light below the diffraction limit. In dielectric materials, light diffraction is a result of the uncertainty principle: limitations to the value of the refractive index of the material, n, cause restrictions in momentum, k, space ($k = n\omega/c$, where ω is the angular frequency of the light and c is the speed of the light in vacuum), resulting in a spreading of the light in real space. Plasmon waves in metallic waveguides[3] or at metal/dielectric interfaces can overcome this fundamental limit by leveraging the negative permittivity of metals, which arises from the collective inertia of electrons under time-varying electric fields, and is usually in the near-infrared to visible frequency range. From the perspective of circuit analysis, this negative but real permittivity corresponds to an imaginary conductivity, revealing the inductive character of the metal. Such an inductance is called 'kinetic inductance', to differentiate it from the traditional 'magnetic inductance', which results from a magnetic field around a current-carrying conductor. For normal metals, the kinetic inductance can be safely ignored at microwave frequencies because their resistance dominates the impedance at this relatively low-frequency range. In superconductors, the kinetic inductance can be significant even at microwave frequencies due to their ideal conductive properties[4].

Writing in *Nature Nanotechnology*, Donhee Ham, Philip Kim and colleagues at Harvard University, Columbia University