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Transient Spin Density Wave Order Induced in the Normal State of $BaFe_2As_2$ by Coherent Lattice Oscillations

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Abstract. We trace the ultrafast dynamics of the spin density wave gap of the pnictide system $BaFe_2As_2$ probing resonantly with broadband multi-terahertz pulses. The photoexcitation in the low-temperature ground state leads to a fast suppression of the spin order followed by a slower recovery process. Surprisingly, in the normal state, we observe periodic oscillations of the spin density wave feature at a frequency as high as 5.5 THz. Our results indicate a transient development of a macroscopic order driven by a coherent lattice oscillation and attest to a pronounced spin–phonon coupling in pnictides.

In recent years, the iron-based pnictides have been established as a new class of high-temperature superconductors. The superconducting state in these materials is achieved by chemical doping or application of external pressure to a parent compound with a spin density wave (SDW) ground state. Therefore, the underlying magnetic order has been at the heart of the discussion concerning the mechanism of high-temperature superconductivity in pnictides [1]. On the other hand, detailed theoretical and experimental studies have pointed out the intimate connection between the lattice structure and the magnetic ordering [2,3]. Therefore, unveiling the interplay among the lattice structure, magnetism and superconductivity is crucial for an understanding of the nature of high-temperature superconductivity in pnictides.

Ultrafast pump-probe experiments provide access to the dynamics of various quantum degrees of freedom after perturbation by ultrashort optical pulses. Thus, direct information about the interplay between single-particle electronic states, collective modes, magnetization and lattice structure may be obtained. The time-resolved multi-THz techniques extending over the far- and mid-infrared spectral ranges with time resolution of a few tens of femtoseconds are particularly useful for resonant probing of single-particle and collective low-energy excitations in complex materials [4].

Here, we employ few-cycle multi-THz pulses to resonantly probe the evolution of the SDW gap of $BaFe_2As_2$ after excitation with a femtosecond optical pulse. This material represents the parent

compound for the Ba-122 family of pnictides - one of the most studied iron-based superconductors due to the availability of large and high-quality single crystals. The $BaFe_2As_2$ samples used in the experiment exhibit a SDW transition at $T_{SDW} = 120$ K. The time-dependent optical conductivity spectra of $BaFe_2As_2$ after photoexcitation by a 12 fs near-infrared pump pulse are measured over the photon energies between 41 meV and 110 meV with a time resolution of 40 fs.

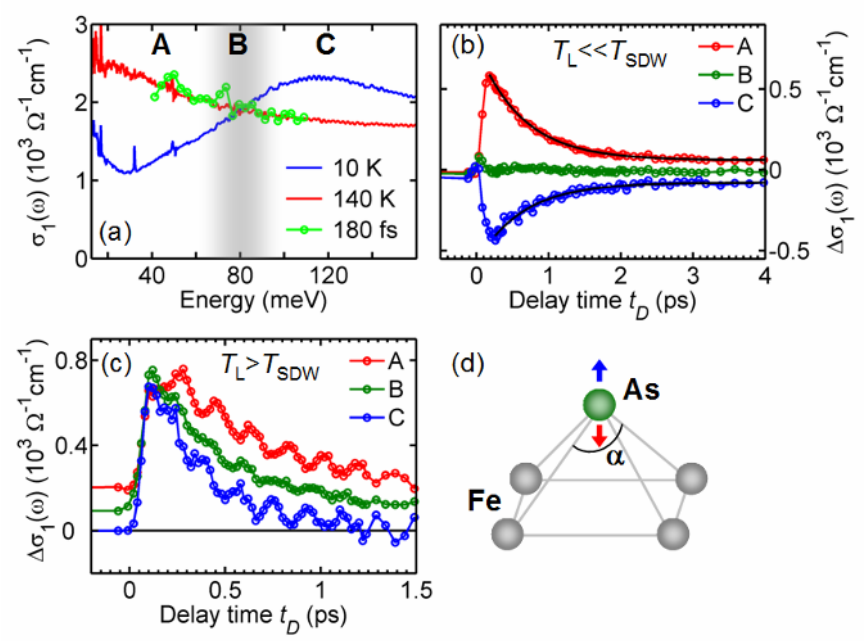

Fig. 1. (a) Equilibrium optical conductivity in the spin density waves state at 10 K (blue line) and in the normal state at 140 K (red line). The conductivity of the excited state 180 fs after optical pumping is shown by green circles. The grey shaded area indicates the spectral region B around the isosbestic point. (b) Conductivity changes spectrally integrated over regions A, B and C, respectively, as shown in panel (a). The black solid lines represent fits to the data with a single exponential decay time. (c) Spectrally averaged conductivity changes in the normal state slightly above T_{SDW}: T_L = 134 K and $\Phi = 530$ μJ/cm^2. The curves are vertically shifted for clarity. (d) Schematic visualization of the A_{1g} phonon vibration at 5.5 THz which involves the motion of arsenic ions with respect to the Fe square lattice. The Fe-As-Fe bonding angle α is modulated upon displacement of arsenic ions.

The equilibrium optical conductivity spectra measured by spectroscopic ellipsometry above and below T_{SDW} are shown in Fig. 1(a). The closure of the SDW gap on heating gives rise to a transfer of spectral weight across the isosbestic point at 80 meV corresponding to the gap edge. In the magnetically ordered state at a base temperature of the lattice T_L = 10 K an absorbed pump fluence of Φ = 63 μJ/cm^2 results in a complete melting of the SDW order reached within 180 fs. After this delay time the transient optical conductivity shown by green circles in Fig. 1(a) agrees well with the equilibrium optical response of the normal state. The melting process is followed by ultrafast recovery of the SDW order with a time constant of 0.63 ps as shown in Fig. 1(b). The optical conductivity changes are averaged in three distinct spectral regions below (A), around (B) and above (C) the gap edge. The opposite signs of the signals in the regions A and C and an almost vanishing signal in region B clearly demonstrate the ultrafast transfer of the spectral weight and, thus, confirm that the observed spectral changes are related to the dynamics of the SDW order.

Fig. 1(c) shows the temporal changes of the averaged optical conductivity in the normal state recorded at $T_L = 134$ K and a higher excitation fluence of Φ = 530 μJ/cm^2. Instead of the strong

dispersion related to the gap closure in the SDW state, the conductivity in the normal state exhibits an overall increase followed by the fast exponential decay. In addition, clear oscillations at a frequency of 5.5 THz are observed. This frequency matches the one of A_{1g} optical phonon modes which involves the displacement of the arsenic ions perpendicular to the square lattice of Fe ions as schematically illustrated in Fig. 1(d).

Most importantly, we find that the observed coherent oscillations are strongly dependent on the probe photon energy. As illustrated in Fig. 1(c), the oscillations in spectral regions A and C are out of phase with each other while the oscillatory response is strongly suppressed in region B. This behavior closely resembles the dispersive signature of the SDW gap observed below T_{SDW} (see Fig. 1(b)). A quantitative analysis of the oscillatory component reveals that it matches the change in the optical conductivity that is induced in the equilibrium state by the development of the SDW gap [5]. This fact represents the most important and surprising result of our study. It indicates that even in the normal state well above T_{SDW} the coherent A_{1g} phonon oscillations periodically induce the transient SDW order.

A qualitative picture of the observed phenomenon can be obtained by considering the impact of the lattice vibration on the electronic band structure of $BaFe_2As_2$. The distance of As from the Fe square lattice modulated by the A_{1g} vibration defines the Fe-As-Fe bonding angle α as shown in Fig. 1(d). This structural parameter is known to significantly affect the Fermi surface nesting in $BaFe_2As_2$ [6]. Furthermore, frozen phonon calculations have demonstrated that a small vibrational displacement of arsenic ions can result in an appreciable modification of the band structure around the Fermi level which gets further enhanced by the magnetic ordering [7]. Thus, the A_{1g} vibration can modify the nesting conditions between the electron and hole Fermi pockets which in turn can induce the SDW order. Remarkably, our findings suggest that the transient SDW develops on a very short time scale and quasi-adiabatically follows the coherent oscillation at a frequency as high as 5.5 THz.

In conclusion, we observe an induction of the transient SDW order in $BaFe_2As_2$ induced by coherent lattice oscillations on a femtosecond time scale. This phenomenon represents a qualitatively new form of coherent control in solids with strong electronic correlations. Our findings attest to a strong impact of coherent atomic motion on magnetic ordering in iron-based pnictides. Therefore, spin-phonon coupling may be considered as a significant factor in the microscopic theory of superconductivity in these materials.

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