Electron-phonon coupling and the charge gap of spin-density wave iron-pnictide materials from quasiparticle relaxation dynamics

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We investigate the quasiparticle relaxation and low-energy electronic structure in undoped $SrFe_2As_2$ exhibiting spin-density wave (SDW) ordering using optical pump-probe femtosecond spectroscopy. A remarkable critical slowing down of the quasiparticle relaxation dynamics at the SDW transition temperature $T_{SDW}=200$ K is observed. From temperature dependence of the transient reflectivity amplitude we determine the SDW-state charge gap magnitude, $2\Delta_{SDW}/k_BT_{SDW}=7.2\pm1$. The second moment of the Eliashberg function, $\lambda\langle(\hbar\omega)^2\rangle=110\pm10$ meV^2, determined from the relaxation time above T_{SDW}, is similar to SmFeAsO and $BaFe_2As_2$ indicating a moderate electron phonon coupling.

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The discovery of high-temperature superconductivity in iron-based pnictides[1–3] has attracted a great deal of attention recently. The question of the relative importance of the lattice and spin degrees of freedom for the superconducting pairing interaction becomes immediately apparent since the superconductivity appears upon doping the parent materials[4] which show the spin-density wave (SDW) ground state. Understanding the parent SDW compounds also from the point of electron-phonon, and not only the spin-spin and spin-charge interactions, is therefore beneficial for understanding the nature of the superconducting coupling in the doped compounds.

Time-resolved spectroscopy has been very instrumental in elucidating the nature of the electronic excitations in superconductors, particularly cuprates[5–19] and recently also iron pnictides.[20–24] Moreover, the relaxation kinetics can give us valuable information on the electronic structure[6] and electron-phonon coupling.[25]

In this work we present a time-resolved femtosecond spectroscopy study of $SrFe_2As_2$ in the normal and the SDW state. From the photoexcited carrier relaxation dynamics we determine the electron-phonon coupling parameters and the charge gap magnitude. We compare the results with recent data[24] in SmFeAsO and find that they are similar both in the SDW and normal state with some minor differences in the magnitude of the response at high temperatures.

Optical experiments were performed using the standard pump-probe technique, with 50 fs optical pulses from a 250-kHz $Ti:Al_2O_3$ regenerative amplifier seeded with an $Ti:Al_2O_3$ oscillator. We used the pump photons with doubled ($\hbar\omega_P=3.1$ eV) photon energy and the probe photons with 1.55 eV photon energy. The pump and probe polarizations were perpendicular to each other and oriented with respect to the crystals to obtain the maximum amplitude of the response at low temperatures. The pump and probe beam diameters were determined by measuring the transmittance of calibrated pinholes mounted at the sample position.[26] Single crystals of $SrFe_2As_2$ were prepared by the self-flux method.[27] For optical measurements the cleaved crystals were glued on a Cu plate mounted in an optical liquid-He flow cryostat.

In Fig. 1 we plot the temperature dependence of $\Delta R/R$ transients in $SrFe_2As_2$. Below T_{SDW} the transients are dominated by the initial single exponential relaxation followed by a weak structure at around 10 ps [see Fig. 1(b)]. At T_{SDW} a critical slowing down of relaxation is observed in the form of a long-lived relaxation tail which is following the initial ~1 ps exponential decay. Above T_{SDW} the amplitude of the initial subpicosecond relaxation strongly drops and the structure on a longer time scale becomes apparent. The behavior is similar to SDW SmFeAsO (Ref. 24) with the exception of the subpicosecond relaxation amplitude above T_{SDW} being smaller in $SrFe_2As_2$.

The amplitude of the initial subpicosecond peak shows a minor departure from the linear pump fluence ($\mathcal{F}$) dependence at the highest $\mathcal{F}$ (see Figs. 2 and 3) while the subpicosecond relaxation time is virtually $\mathcal{F}$ independent (see Fig. 3). At low T however, an additional nonexponential slow relaxation component appears at the lowest fluence (see Fig. 2) which can be (beyond 5–10 ps) attributed to the heat diffusion out of the excitation volume[28] as indicated by fits in Fig. 2.

In a metal the photoexcited-quasiparticle relaxation time is governed by transfer of energy from electronic degrees of freedom to lattice degrees of freedom. Recently the problem was solved analytically.[25] In bad metals, above the Debye frequency (ω_D), the relaxation time linearly depends on the temperature, T, where the slope is determined by the inverse of the second moment of the Eliashberg function $\lambda\langle\omega^2\rangle$ (Refs. 25 and 29)

$$\tau=\frac{2\pi k_BT}{3\hbar\lambda\langle\omega^2\rangle}. \quad (1)$$

We find that above ~230 K our τ data nicely follow the predicted linear T dependence [see Fig. 4(a)] with $\lambda\langle(\hbar\omega)^2\rangle=110\pm10$ meV^2. The phonon spectrum of $SrFe_2As_2$ extends up to ~40 meV with the acoustic phonon cutoff at ~10 meV.[30] To estimate the electron-phonon coupling constant, λ, we determine the ratio $\lambda\langle(\hbar\omega)^2\rangle/\lambda$ by assuming that the electron-phonon spectral function, $\alpha^2F(\omega)$, has the phonon density-of-states (DOS) shape[30,31] and obtain. $\lambda\langle(\hbar\omega)^2\rangle/\lambda\simeq430$ meV^2. This gives $\lambda\simeq0.25$, which is

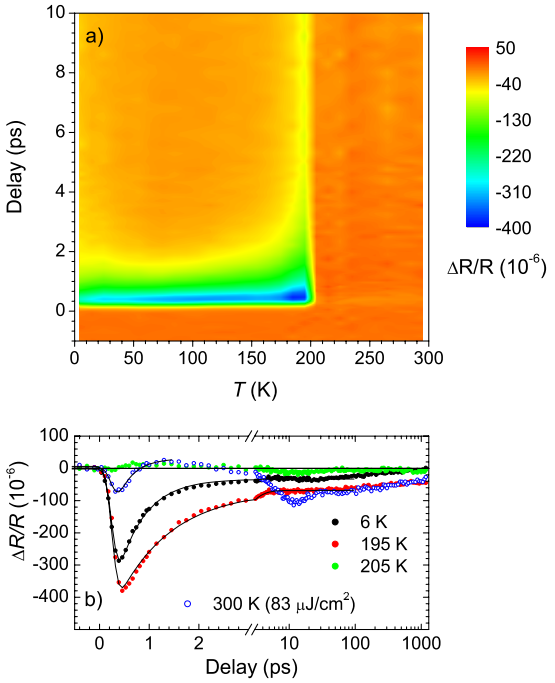

FIG. 1. (Color online) (a) $\Delta R/R$ transients as a function of temperature at pump fluence 17 $\mu J/cm^2$. (b) $\Delta R/R$ transients from (a) at representative temperatures with single-exponential decay fits. A divergent relaxation time at $T_{SDW}=200$ K is clearly seen in (a).

rather low to explain the superconducting critical temperatures in doped compounds within a standard BCS model.[31] If however, $\alpha^2F(\omega)$ is for some reason enhanced in the low-energy phonons region, λ could easily reach significantly higher values.

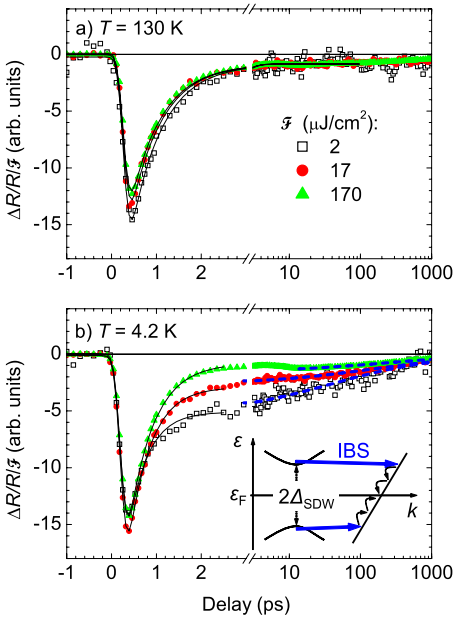

FIG. 2. (Color online) Fluence dependence of normalized $\Delta R/R$ transients in the SDW state. The black thin lies are single exponential decay fits and blue dashed lines the one-dimensional diffusion (Ref. 28) fits. In the inset to (b) the relaxation pathway via interband scattering, that is discussed in text, is schematically shown.

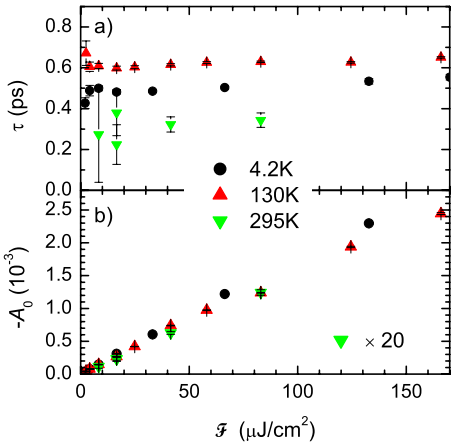

FIG. 3. (Color online) Fluence dependence of the $\Delta R/R$ transient amplitude and relaxation time at different temperatures.

Owing to the multiband nature of iron pnictides it is possible, that due to optical selection rules some bands are not directly detected in $\Delta R/R$ transients. However, at the temperatures of interest the momentum (interband) scattering is expected to be strong resulting in effective averaging of $\lambda\langle(\hbar\omega)^2\rangle$ over all the bands.

Below T_{SDW} the increasing amplitude of the subpicosecond transients indicates the appearance of a bottleneck in the photoexcited electron relaxation. The bottleneck is associated with opening of a T-dependent charge gap[6] due to SDW formation and concurrent Fermi surface reconstruction.[32,33] We use Eq. (6) from Kabanov *et al.*[6]

$$\Delta R/R \propto n_{pe} \propto 1/[\Delta(T) + k_B T/2]\{1 + g\sqrt{k_B T/\Delta(T)} \times \exp[-k_B T/\Delta(T)]\}, \quad (2)$$

which describes the photoexcited change in quasiparticle density in the presence of a temperature-dependent gap, $\Delta(T)$, to fit the amplitude T dependence below $T_{SDW}=200$ K. The parameter g represents the ratio between the relevant bosonic and electronic density of states. Relation (2)

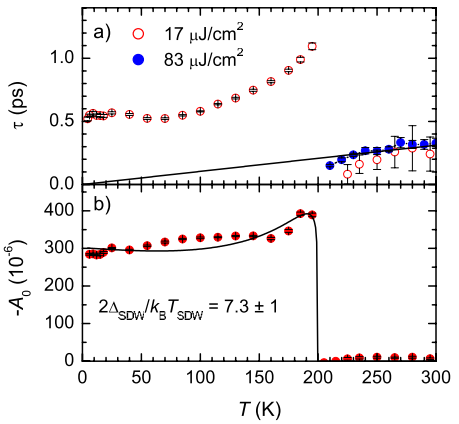

FIG. 4. (Color online) (a) The relaxation time at two pump fluences and (b) amplitude at $\mathcal{F}=17$ $\mu J/cm^2$ as functions of temperature. The black solid line in (a) is fit of Eq. (1) to τ above 230 K. The black solid line in (b) represents the fit of Eq. (2).

TABLE I. Electron phonon coupling parameters and SDW gap magnitudes in SDW iron pnictides. The experimental inelastic-neutron-scattering phonon spectra from Mittal *et al.* (Refs. 30 and 40) and Osborn *et al.* (Ref. 41) were used to estimate $\lambda\langle(\hbar\omega)^2\rangle/\lambda$ assuming $\alpha^2F(\omega)$ has the phonon DOS shape.

	$\lambda\langle(\hbar\omega)^2\rangle$ (meV^2)	$\lambda\langle(\hbar\omega)^2\rangle/\lambda$ (meV^2)	λ	$2\Delta_{SDW}/k_BT_{SDW}$
$SrFe_2As_2$	110 ± 10	430	~0.25	7.2 ± 1
SmFeAsO (Ref. 24)	135 ± 10	770	~0.18	~5
$BaFe_2As_2$ [a]	~65 [b]	430	~0.15	4.7 ± 1.6

[a]We used the data from the supplemental material of Chia *et al.* (Ref. 22).
[b]Only data just above $T_{SDW}\simeq130$ K is available so the value might be underestimated.

was initially derived[6] for the case of a momentum independent gap. The assumption behind Eq. (2) is that at a certain time scale the bosonic spectrum can be divided in the low and the high energy parts at the characteristic energy $2\Delta(T)$ determined by the structure of the electronic density of states. During the relaxation a temporary quasiequilibrium is achieved between the high-energy bosonic and electronic degrees of freedom resulting in the characteristic T dependence in Eq. (2). Since the underlying assumption is rather general the use of relation (2) can be extended to the momentum-dependent gap case. In this case $2\Delta(T)$ represents a typical bosonic energy involved in the relaxation, which is directly related to the characteristic energy scale of the gap.

Using a single $\Delta_{SDW}(T)$ with the BCS temperature dependence and $2\Delta_{SDW}/k_BT_{SDW}\simeq7.2\pm1$ results in a rather good fit to the amplitude temperature dependence [see Fig. 4(b)]. The observed characteristic gap energy is close to the magnitude of the largest of the two SDW gaps obtained from the optical conductivity.[34] Since the interband scattering is strong, as argued below, the smallest of the gaps should present the bottleneck for the hot carrier energy relaxation so our data do not confirm the existence of the smaller gap suggested by Hu *et al.*[34]

Similarly to the case of SmFeAsO (Ref. 24) we observe no decrease of the relaxation time with $\mathcal{F}$ as predicted by Kabanov *et al.*[6] We also observe no divergence of the relaxation time with decreasing T such as in heavy fermion SDW $UNiGa_5$,[35] which, similar to $SrFe_2As_2$,[36] remains metallic[37] upon the SDW gap opening. We can rule out the ballistic hot electron escape as a source of the low-T relaxation-time divergence cutoff[38] due to the relatively high resistivity of $SrFe_2As_2$ (Ref. 36) and large optical penetration depth of ~60 nm at $\hbar\omega_{probe}=1.55$ eV.[39] However, in iron pnictides, due to the presence of ungapped electronic bands below T_{SDW}, the energy relaxation is not limited by the anharmonic energy transfer from the high-frequency to the low-frequency phonons, but rather by the interband scattering (IBS) from the states at the edge of the SDW gap to the states in ungapped band(s) with energies $\epsilon-\epsilon_F\gg k_BT$ [see inset to Fig. 2]. Such scattering can be enhanced by the presence of impurities, which may explain the difference between the higher residual resistivity in $SrFe_2As_2$ and lower residual resistivity in $UNiGa_5$.

The absence of multiple relaxation components (apart from diffusion) from the $\Delta R/R$ transients together with IBS imply that mainly the hot carriers from the SDW gaped electronic bands contribute to the photoinduced reflectivity transients. This does not hinder the determination of $\lambda\langle(\hbar\omega)^2\rangle$ as discussed above since the momentum scattering, which becomes faster than the energy relaxation rate at high temperatures, results in an effective averaging of $\lambda\langle(\hbar\omega)^2\rangle$ over all the bands.

In conclusion, we compare in Table I our results in $SrFe_2As_2$ to SmFeAsO (Ref. 24) and $BaFe_2As_2$.[22] In all three compounds the second moment of the Eliashberg function is moderate suggesting similar values of the electron phonon coupling constant λ as suggested by the density-functional theory using linear response,[31] unless $\alpha^2F(\omega)$ is strongly enhanced in the low-energy phonons region, contrary to the density-functional-theory prediction.[31] Similarly in all three compounds the temperature dependence of the relaxation below T_{SDW} indicates the appearance of a quasiparticle relaxation bottleneck due to opening of a charge gap at T_{SDW} with a BCS-type temperature dependence.

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