Electron-phonon coupling and charge gap in spin-density-wave iron-pnictides from quasiparticle relaxation dynamics

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We investigate the quasiparticle relaxation and low-energy electronic structure in undoped SrFe2As2 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, $2\Delta_{SDW}/k_B T_{SDW} = 7.2 \pm 1$. The second moment of the Eliashberg function, $\lambda\langle(h\omega)^2\rangle = 110 \pm 10$ meV$, determined from the relaxation time above $T_{SDW}$, is similar to SmFeAsO and BaFe$_2$As$_2$ indicating a rather small electron phonon coupling constant unless the electron-phonon spectral function $(\alpha^2 F(\omega))$ is strongly enhanced in the low-energy phonon region.

The discovery of high-temperature superconductivity in iron-based pnictides [11–13] 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 [14–19], which show the spin-density wave (SDW) ground state. Understanding the parent SDW compounds 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 [4–18] and recently also iron pnictides [19–23]. Moreover, the relaxation kinetics can give us valuable information on the electronic structure [2] and electron-phonon coupling [24].

In this work we present a time-resolved femtosecond spectroscopy study of SrFe$_2$As$_2$ in the normal and the SDW state. From the photo-excited carrier relaxation dynamics we determine the electron-phonon coupling parameters and the charge gap magnitude. We compare the results with recent data [22] 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$_2$O$_3$ regenerative amplifier seeded with an Ti:Al$_2$O$_3$ oscillator. We used the pump photons with doubled ($h\nu_P = 3.1$ 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 [24]. Single crystals of SrFe$_2$As$_2$ were prepared by the self-flux method [26]. 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$_2$As$_2$. Below $T_{SDW}$ the transients are dominated by the initial single exponential relaxation followed by a weak structure at around 10 ps (see Fig. 2 (a)). At $T_{SDW}$ a critical slowing down of relaxation is observed in the form of a long lived relaxation which is following the initial $\sim 1$ ps exponential decay. Above $T_{SDW}$ the amplitude of the initial sub-ps relaxation strongly drops and the structure on a longer timescale becomes apparent. The behavior is similar to SDW SmFeAsO [22] with the exception of the sub-ps relaxation amplitude above $T_{SDW}$ being smaller in SrFe$_2$As$_2$.

The amplitude of the initial sub-ps peak shows a minor departure from the linear pump fluence ($F$) dependence at the highest $F$ (see Figs 3 and 4) while the sub-ps relaxation time is virtually $F$ independent (see Fig. 4). At low $T$ however, an additional non-exponential slow relaxation component appears at the lowest fluence (see Fig. 3) which can be (beyond 5-10 ps) attributed to the heat diffusion out of the excitation volume [27] as indicated by...
fits in Fig. 3.

In a metal the photoexcited-quasiparticle relaxation time is governed by transfer of energy from electronic degrees of freedom to the lattice degrees of freedom. Recently the problem was solved analytically\cite{24}. In bad metals, above the Debye frequency ($\omega_D$), 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 $\lambda(\omega^2)$: $\tau = \frac{2 \pi k_B T}{3 \hbar \lambda(\omega^2)}$. \hfill (1)

We find that above $\sim 230$ K our $\tau$-data nicely follow the predicted linear $T$ dependence (see Fig. 2 (b)) with $\lambda(\hbar \omega^2) = 110 \pm 10$ meV. The phonon spectrum of SrFe$_2$As$_2$ extends up to $\sim 40$ meV with the acoustic phonon cutoff at $\sim 10$ meV.\cite{29} We determine $\lambda(\hbar \omega^2)/\lambda$ by assuming that the electron-phonon spectral function, $\alpha^2 F(\omega)$, has the phonon DOS shape\cite{29,30} and obtain $\lambda(\hbar \omega^2)/\lambda \approx 430$ meV. This gives $\lambda \approx 0.25$, which is rather low to explain the superconducting critical temperatures in doped compounds within a standard single band BCS model. If however, $\alpha^2 F(\omega)$ is for some reason enhanced in the low-energy phonons region, $\lambda$ could easily reach significantly higher values.

Below $T_{SDW}$ the increasing amplitude of the sub-ps transients indicates the appearance of a bottleneck in the photo-excited electron relaxation. The bottleneck is associated with opening of a $T$-dependent gap due to SDW formation resulting in the Fermi surface reconstruction\cite{31,32}. We use equation (6) from Kabanov et al.\cite{5}, which describes the photo-excited change in quasiparticle density in the presence of a temperature dependent gap, to fit the amplitude below $T_{SDW} = 200$ K. Using a single SDW gap energy with the BCS temperature dependence and $2\Delta_{SDW}/k_B T_{SDW} \approx 7.2 \pm 1$ results in a rather good fit to the amplitude temperature dependence (see Fig. 2 (c)). The observed gap magnitude is close to the magnitude of the largest of the two SDW gaps obtained from the optical conductivity.\cite{33} Since the inter-band scattering is strong, as argued below, the

Figure 2: (Color online) $\Delta R/R$ transients at representative temperatures with single-exponential decay fits (a). The relaxation time at two pump fluences (b) and amplitude at $F = 17 \mu J/cm^2$ (c) as functions of temperature. The black solid line in (b) is fit of equation (1) to $\tau$ above 230 K. The black solid line in (c) represents the fit of equation (6) from\cite{5}.

Figure 3: (Color online) Fluence dependence of normalized $\Delta R/R$ transients in the SDW state. The black thin lines are single exponential decay fits and blue dashed lines the 1D diffusion fits. In the inset to (b) the relaxation pathway via inter-band scattering, that is discussed in text, is schematically shown.
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.\[32\].

Similarly to the case of SmFeAsO\[23\] we observe no decrease of the relaxation time with $T$ as predicted by Kabanov et al.\[5\]. We also observe no divergence of the relaxation time with decreasing $T$ such as in heavy fermion SDW UNiGa$_5$\[35\] which, similar to as SrFe$_2$As$_2$,\[23\] remains metallic\[36\] upon the SDW gap opening. We can rule out the ballistic hot electron escape as a source of the low-$T$ relaxation-time divergence cut-off\[37\] due to the relatively high resistivity of SrFe$_2$As$_2$\[35\] and large optical penetration depth of $\sim$ 60 nm at $\hbar \omega_{\text{probe}} = 1.55$ eV\[14\]. However, in iron pnictides, due to the presence of ungapped electronic bands below $T_{\text{SDW}}$, the energy relaxation is not limited by the anharmonic energy transfer from the high frequency to the low frequency phonons, but rather by the inter-band scattering (IBS) from the states at the edge of the SDW gap to the states in ungapped band(s) with energies $\epsilon - \epsilon_F \gg k_B T$ (see inset to Fig. 3(b)). Such scattering can be enhanced by the presence of impurities, which may explain the difference between the higher residual resistivity SrFe$_2$As$_2$ and lower residual resistivity 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 gap electronic bands contribute to the photoduced reflectivity transients. This does not hinder the determination of $\lambda((\hbar \omega)^2)/\lambda$ at high temperatures, since the IBS is essentially a momentum scattering, which becomes faster than the energy relaxation rate at high temperatures.

By means of the femtosecond optical pump-probe spectroscopy we determined the second moment of the Elishberg function in SDW SrFe$_2$As$_2$ and compared it to SmFeAsO and BaFe$_2$As$_2$. In all compounds the values are similar (see Table I) suggesting moderate values of the electron phonon coupling constant $\lambda$ as suggested by the linear response theory,\[30\] unless $\alpha^2 F(\omega)$ is strongly enhanced in the low-energy phonons region.

Below $T_{\text{SDW}}$ the temperature dependence of the relaxation indicates the appearance of a quasi-particle relaxation bottleneck due to opening of a single charge gap at $T_{\text{SDW}}$ with a BCS-like temperature dependence similar to that observed\[23\] in SmFeAsO and inferred from the recently published data\[38\] in BaFe$_2$As$_2$.

![Figure 4](http://prl.aps.org/supplemental/PRL/v104/i2/e027003).

|                | $\lambda((\hbar \omega)^2)/\lambda$ (meV)$^2$ | $\lambda$ (meV)$^2$ | $2\Delta_{\text{SDW}}/k_B T_{\text{SDW}}$ |
|----------------|-----------------------------------------------|---------------------|------------------------------------------|
| SrFe$_2$As$_2$ | $110 \pm 10$                                  | 430                 | $\sim 0.25$                              | 7.2 $\pm$ 1                                 |
| SmFeAsO ref.\[23\] | $135 \pm 10$                                  | 770                 | $\sim 0.18$                              | 5                                           |
| BaFe$_2$As$_2$ $^a$ | $65^b$                                        | 430                 | $\sim 0.15$                              | 4.7 $\pm$ 1                                 |

$^a$We used the data from the supplemental material of Chia et al.\[38\]. \(\text{http://prl.aps.org/supplemental/PRL/v104/i2/e027003}\).

$^b$Only data just above $T_{\text{SDW}} \approx 130K$ is available so the value might be underestimated.

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.\[21, 33\] and Osborn et al.\[11\] were used to estimate $\lambda((\hbar \omega)^2)/\lambda$ assuming $\alpha^2 F(\omega)$ has the phonon DOS shape.

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