Time-Resolved Quasiparticle Dynamics in the Spin-Density-Wave State

Elbert E. M. Chia, Jian-Xin Zhu, H. J. Lee, Namjung Hur, N. O. Moreno, R. D. Averitt, J. L. Sarrao, and A. J. Taylor
Los Alamos National Laboratory, Los Alamos NM 87545, USA
(Dated: March 11, 2018)

Time-resolved photoinduced reflectivity is measured in the spin-density-wave (SDW) phase using itinerant antiferromagnets UMGa5 (M=Ni, Pt). For UNiGa5 [TN=85 K, Q=(π,π,π)], the relaxation time τ shows a sharp increase at TN consistent with the opening of a SDW gap. For UPtGa5 [TN=26 K, Q=(0,0,π)], no change in τ is observed at TN or at the lowest temperatures. We attribute this to the absence of the SDW gap at the Fermi level, due to a different modulation vector Q, which leads to a gapless quasiparticle spectrum. Our results challenge the conventional wisdom that a SDW phase necessarily implies a SDW gap at the Fermi level.

The quasiparticle (QP) dynamics of the spin-density-wave (SDW) phase is an important area of study, especially in the cuprate high-temperature superconductors (HTSC), where superconductivity and antiferromagnetism (AFM) could coexist, either in an applied magnetic field [1,2], or in zero field [3]. In single-layered non-superconducting cuprates, where there is only a single layer of CuO$_2$ planes in the unit cell, AFM is necessarily G-type, where the nearest-neighbor spins in the CuO$_2$ plane are antiferromagnetically (AF)-aligned. However, in multi-layered cuprates, two types of AFM can occur on the CuO$_2$planes: (1) G-type AFM, or (2) A-type AFM, i.e. spins are ferromagnetically (FM) aligned along each plane, but spins on adjacent planes are AF-aligned. Being able to elucidate the magnetic alignment of the spins in multi-layered cuprate HTSCs in the coexistence phase is crucial in narrowing down the starting point for a pairing theory in the HTSCs. It is therefore important to study the QP dynamics of pure SDW systems before proceeding to more complex coexistence phases in HTSCs. At present there have been no systematic measurements of the QP dynamics of the SDW phase, as a function of the type of AFM, where below TN, a SDW gap, i.e. a charge gap on the nested parts of the Fermi surface (FS), might open up.

Usually, the SDW state in itinerant AFMs is probed by techniques such as resistivity, specific heat and neutron scattering. In resistivity and specific heat, a feature appearing at TN, such as a hump or peak, has been interpreted as due to the formation of a SDW gap ∆SDW and the accompanying partial disappearance of the FS (See, for example, Ref. [4,5,6]. However, it is not clear if the feature at TN is due to an actual gap opening up in the DOS at the Fermi level [DOS(EF)], or merely due to a decrease in DOS(EF) without it vanishing. In elastic neutron scattering (ENS), the intensity of the neutron scattering peak $I_{NS}$ increases from zero below TN in a BCS-like manner, and is proportional to $M^2$, where M is the staggered magnetization. In an itinerant AFM, Overhauser [7] derived $M \propto \Delta_{SDW}$, yielding $\Delta_{SDW} \propto \sqrt{I_{NS}}$. However, does the presence of a staggered magnetization always imply the presence of the gap in the DOS? Looking at this problem from another perspective, does the opening up of a gap depend on the type of AFM in the material?

Ultrafast optical spectroscopy (UOS) has recently been used in the study of correlated electron materials. For example, in heavy fermions (HF) such as YbAgCu$_4$ [8] and CeCoIn$_5$ [9], time-resolved photoinduced reflectivity measurements display a divergence of the electron-phonon (e-ph) relaxation time τ at the lowest temperatures $T$. In materials with a gap in the QP spectrum such as HTSCs like YBCO [10,11] and charge-density-wave materials like K$_{0.3}$MoO$_3$ [12], τ diverges near $T_c$, when a gap opens in the QP density of states (DOS). The $T$ dependence of the relaxation time and peak amplitude has been explained by the phenomenological Rothwarf-Taylor (RT) model. This model describes the relaxation of photoexcited SCs [13], where the presence of a gap in the electronic DOS gives rise to a relaxation bottleneck for carrier relaxation, arising in SCs from the competition between QP recombination and pair breaking by phonons [14].

In this Letter, we investigate the QP response in the SDW state using UMGa5 (M=Ni, Pt) as a model system. For UNiGa5, the decay time of $\Delta R/R$ (directly related to $\tau$) increases sharply at TN and shows a quasi-divergence below TN, consistent with the opening of a SDW gap. For UPtGa5, however, there is no discernible change in $\tau$ across TN and at the lowest $T$. We attribute this to the QP spectrum being gapless, which is the result of the SDW modulation vector Q being different from that of UNiGa5. Our technique thus enables us to distinguish the SDW state of two very similar materials, one with a SDW gap (UNiGa5), and the other without a SDW gap (UPtGa5). We further substantiate our claim by performing a microscopic model calculation on a 3-dimensional (3D) cubic lattice in the SDW state, and show that the presence or absence of a SDW gap is intimately related to the value of Q of that particular material. Our results challenge the conventional wisdom that a SDW phase necessarily implies a SDW gap at the Fermi level.

UMGa5 (M=Ni, Pt) are 5f itinerant AFMs with
$T_N \approx 85$ K (Ni) and 26 K (Pt), respectively. The AF phase in UNiGa$_5$ is G-type, i.e. the nearest-neighbor spins are AF-aligned, with modulation vector $Q = (\pi, \pi, \pi)$ \cite{15}. Contrast this with UPtGa$_5$, where the AF phase is A-type, i.e. the spins are FM-aligned in the $ab$ plane and AF along the $c$ axis, with $Q = (0,0,\pi)$ \cite{14}. The electronic specific heat coefficients $\gamma = 30$ mJ/mol.K$^2$ (Ni), and 57 mJ/mol.K$^2$ (Pt), respectively \cite{14,15}. The moderate values of $\gamma$ suggest that these materials are marginal HFs.

Single crystals of UMGa$_5$ were grown in Ga flux \cite{16}, with dimensions $\sim 1 \times 1 \times 0.4$mm$^3$. Specific heat measurements were performed in a Quantum Design PPMS from 2 K to 300 K to determine $T_N$. The shape of the specific heat anomaly at $T_N$ is similar for both materials. The photoinduced reflectivity measurements were performed using a standard pump-probe technique \cite{17}, with a Ti:sapphire laser producing sub-100 fs pulses at approximately 800 nm (1.5 eV) as the source of both pump and probe optical pulses. The pump and probe pulses were cross-polarized. The experiments were performed with a pump fluence of $< 1.0 \mu$J/cm$^2$, yielding a photoexcited QP density $n_{pc} < 0.05$ unit cell. The probe intensity was $\sim 25$ times lower. Data were taken from 10 K to 300 K. The photoinduced $T$ rise at the lowest $T$ was estimated to be $\sim 10$ K for UNiGa$_5$ and $\sim 6$ K for UPtGa$_5$ (accounted for in all data plots).

In Figure 1 we show the time dependence of the photoinduced signal of UNiGa$_5$ and UPtGa$_5$ below and above $T_N$. The time evolution of the photoinduced reflection $\Delta R/R$ first shows a rapid rise time (of the order of the pump pulse duration) followed by a subsequent picosecond decay. These data can be fit using a single exponential decay over the entire $T$ range, $\Delta R/R = A \exp(-t/\tau)$.

Figure 2 shows the $T$ dependence of the relaxation time $\tau$ for UNiGa$_5$ and UPtGa$_5$ extracted from Fig. 1. For UNiGa$_5$ (solid circles) $\tau$ shows an abrupt increase near $T_N$ followed by a gradual increase at the lowest $T$. The QP dynamics of $\tau$ has been explained by us in Ref. 18 using the RT model, where the opening of the SDW gap, which is the charge gap that opens up along the nested regions of the Fermi surface (FS), leads to a relaxation bottleneck. In contrast, for UPtGa$_5$ (open circles), there is no discernible change in $\tau$ across $T_N$ or at the lowest $T$, implying that no SDW gap opens up in the AF phase. In fact, this $T$ dependence is similar to what has been observed in conventional wide-band metals \cite{14}.

We propose a theoretical model below to explain why a SDW gap opens up in UNiGa$_5$ but not in UPtGa$_5$. We attribute the absence of a SDW gap to be a direct consequence of the value of the AF modulation vector $Q = (0,0,\pi)$ in UPtGa$_5$, compared with $(\pi,\pi,\pi)$ in UNiGa$_5$. The model Hamiltonian for the SDW can be

![Image](image-url)
expressed as

\[ \mathcal{H} = \sum_{k, \sigma} \epsilon_k \hat{c}_k^\dagger \hat{c}_k + \frac{U}{N} \sum_{k, k'} \hat{c}_{k_1}^\dagger \hat{c}_{k_2} \hat{c}_{k_1 + Q}^\dagger \hat{c}_{k_2 + Q} \]  

(1)

where \( \hat{c}_{k, \sigma}^\dagger \) is the creation (annihilation) operator of an electron having the wavenumber \( k \) and spin \( \sigma \). The first term represents the one-electron energy with dispersion \( \epsilon_k \), which, for a 3D cubic lattice, is given by

\[ \epsilon_k = -2t(\cos k_x a + \cos k_y a + \cos k_z a) \]  

(2)

where \( t \) is the overlap integral between the 5f wavefunctions on neighboring sites, and \( a \) is the lattice spacing. The second term in \( \mathcal{H} \) denotes the on-site Coulomb interaction, with \( \mathcal{U} \) being the on-site Coulomb energy, and \( N \) the number of lattice sites.

We define the SDW order parameter \( M_Q \)

\[ M_Q = \frac{-U}{N} \sum_k \langle c_{k+Q}^\dagger c_k \rangle. \]  

(3)

Then, after performing unitary transformation, bilinearization and diagonalization, we obtain the \( T \) dependence of \( M_Q \) in the following self-consistent equation

\[ M_Q = \frac{U}{N} \sum_{k \in rBZ} \left\{ \frac{-M_Q [f(E_+) - f(E_-)]}{\sqrt{\frac{1}{4}(\epsilon_k - \epsilon_{k+Q})^2 + M_Q^2}} \right\} \]  

(4)

where \( f \) is the Fermi function, the \( k \)-points are taken from the reduced Brillouin Zone (rBZ), and \( E_+ \) and \( E_- \) are the two branches of the one-particle energy in the SDW state given by

\[ E_\pm(k) = \frac{1}{2}(\epsilon_k + \epsilon_{k+Q}) \pm \frac{1}{2}\sqrt{4(\epsilon_k - \epsilon_{k+Q})^2 + 4M_Q^2}. \]  

(5)

Fig. 3 shows the \( T \) dependence of \( M_Q \) for \( Q=(\pi, \pi, \pi) \) (solid circles) and \( Q=(0,0,\pi) \) (solid squares). Experimental ENS intensity for (+) UPtGa, and (×) UNiGa, taken from Ref. 16.

Fig. 4 shows the \( T \) dependence of the DOS in the SDW state for (a) \( Q=(\pi, \pi, \pi) \), and (b) \( Q=(0,0,\pi) \). The curves A–G represents DOS calculated from the values of \( T \) and \( M_Q(T) \) in Fig. 3. \( E=0 \) is the Fermi level.
in Eq. [1] $f'$ becomes the delta-function in the $T=0$ limit. The resulting DOS is shown in Fig. 2 where the letters A–G represents the calculated DOS based on the values of $T$ and $M_Q(T)$ in Fig. 3. The Fermi level is located at $E=0$. For a $Q=(\pi, \pi, \pi)$ SDW material, illustrated in Fig. 3, a well-defined energy gap develops below $T_N$, as evidenced by the sharp dips and coherence peaks appearing at $\pm M_Q$. With decreasing $T$, the dips approach zero, and the gaps are more well-defined due to less thermal smearing. For $Q=(0,0,\pi)$, however, though dips do appear below $T_N$, the magnitude of the dips were very small ($\sim 10^{-3}$), even near $T=0$. In the same scale as Fig. 3, it is almost flat, i.e. energy-independent, as shown in the inset of Fig. 4.

We therefore conclude that in UNiGa$_5$ [$Q=(\pi, \pi, \pi)$], a well-defined energy gap forms near the Fermi level in the SDW phase, whereas in UPtGa$_5$ [$Q=(0,0,\pi)$], DOS is finite and almost constant below and above the Fermi level. This therefore agrees with our experimental results, where, in the SDW phase below $T_N$, we see a charge gap opening up in UNiGa$_5$ but not in UPtGa$_5$.

Two pieces of experimental data might suggest the presence of a gap in the QP spectrum in UPtGa$_5$, in contradiction to our results. First, resistivity data $\rho(T)$ showed a small hump at $T_N$, similar to UNiGa$_5$, which the authors attributed to an opening of a gap in the band structure and the partial disappearance of the FS. However, closer examination reveals that for UPtGa$_5$, for both current directions [100] and [001], $\rho$ did not increase upon entering the AF phase, unlike in UNiGa$_5$, where in the [001] direction, $\rho$ increases just below $T_N$, which is identical to the behavior of Cr upon entering the AF phase. The hump seen in UPtGa$_5$ could be merely due to a slight decrease in the QP DOS below $T_N$, as shown in our analysis in Fig. 4, without actually forming a gap on the FS. We also emphasize that the low-$T$ ($T < T_N$) $\rho(T)$ data in Ref. 14 yields a value of $\Delta$ that is not the SDW gap, but is rather the spin gap, i.e. a gap in the magnon dispersion. Second, ENS intensity $I_{NS}$ increases below $T_N$ in a BCS-like manner, which may suggest that a SDW gap $\Delta$ also opens up in the AF phase, since $\Delta \propto \sqrt{T_N}$. However, this expression strictly only applies to G-type AF phase, where the nearest-neighbor spins are AF-aligned as in UNiGa$_5$. We have shown in our previous analysis that $\Delta \propto \sqrt{T_N}$ is not true in the A-type AF state, where the spins are FM-aligned in the $ab$ plane and AF-aligned along the $c$ axis, as in UPtGa$_5$. $I_{NS}(T)$ from ENS merely measures the order parameter in the SDW phase, i.e. the staggered magnetization $M_Q$. It strictly does not measure the SDW gap. Hence our results do not contradict other experimental results. According to the RT model, the lack of a gap at the FS also explains why there is no upturn in $\tau$ at the lowest $T$. Hence we have shown that due to a different modulation vector $Q$ in UPtGa$_5$, a gap does not open up at the Fermi level, resulting in a lack of upturn in $\tau$ at both $T_N$ and at the lowest $T$.

It is interesting to note from Fig. 2 that the value of $\tau$ for UPtGa$_5$ is sub-ps, and is almost $T$-independent. This is commonly seen in metals, where the comparable electron-electron (e-e) and e-ph relaxation rates results in the electron gas not being able to reach thermal equilibrium long before e-ph energy relaxation process sets in. An initial non-thermal electron distribution is thus a necessary starting point to derive the $T$-independence and the sub-ps value of $\tau$. Since both UNiGa$_5$ and UPtGa$_5$ are good metals, the value of their $\tau$’s in the paramagnetic (PM) phase, the $T$-independence of $\tau$ of UPtGa$_5$, as well as their similar values of DOS in the PM phase (from Fig. 2), are consistent with a non-thermal electron distribution immediately following photoexcitation.

We have performed time-resolved photoinduced reflectivity measurements in the SDW phase using itinerant AFMs UMGa$_5$ (M=Ni, Pt) as model systems. For UNiGa$_5$ [$T_N=85$ K, $Q=(\pi, \pi, \pi)$], the relaxation time $\tau$ shows a sharp increase at $T_N$ consistent with the opening of a SDW gap. For UPtGa$_5$ [$T_N=26$ K, $Q=(0,0,\pi)$], no change in $\tau$ was seen at both $T_N$ and at the lowest $T$. We attribute this to the absence of the SDW gap at the Fermi level, due to a different modulation vector $Q$, which leads to a gapless QP spectrum. Our analysis also applies to 2D materials. Our study thus extends the utility of UOS to study SDW materials, that enables us to sensitively probe the presence or absence of a SDW gap in the AF phase.

Work at Los Alamos was supported by the Los Alamos LDRD program. E.E.M.C. acknowledges G. T. Seaborg Postdoctoral Fellowship support.

* Department of Physics, University of California at Berkeley, California, USA
1 Department of Physics, Inha University, Incheon 402-751, South Korea
2 Department of Physics, Federal University of Sergipe, São Cristóvão, SE 49100-000, Brazil
3 B. Lake et al., Nature 415, 299 (2002).
4 H. J. Kang et al., Nature 423, 522 (2003).
5 H. Mukuda et al., Phys. Rev. Lett. 96, 087001 (2006).
6 Y. Tokiwa et al., J. Phys. Soc. Jpn. 70, 1744 (2001).
7 Y. Tokiwa et al., J. Phys. Soc. Jpn. 71, 845 (2002).
8 A. W. Overhauser, Phys. Rev. 128, 1437 (1962).
9 J. Demsar et al., Phys. Rev. Lett. 91, 027401 (2003).
10 J. Demsar, J. L. Sarrao, and A. J. Taylor, J. Phys.: Condens. Matter 18, R281 (2006).
11 S. G. Han et al., Phys. Rev. Lett. 65, 2708 (1990).
12 J. Demsar et al., Europhys. Lett. 45, 381 (1999).
13 J. Demsar, K. Biljakovic, and D. Mihailovic, Phys. Rev. Lett. 83, 800 (1999).
14 J. Demsar et al., Phys. Rev. Lett. 91, 267002 (2003).
15 Y. Tokiwa et al., J. Phys. Soc. Jpn. 71, 725 (2002).
[16] N. O. Moreno et al., Phys. Rev. B 72, 035119 (2005).
[17] D. Mihailovic and J. Demsar, in Spectroscopy of Superconducting Materials, ACS Symposium Series 730, edited by E. Faulques (The American Chemical Society, Washington D. C., 1999), p. 230.
[18] E. E. M. Chia et al., To be published in Phys. Rev. B (Rapid Communications) (2006).
[19] R. H. M. Groeneveld, R. Sprik, and A. Lagendijk, Phys. Rev. B 51, 11433 (1995).
[20] N. H. Andersen and H. Smith, Phys. Rev. B 19, 384 (1979).