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A THEORETICAL STUDY OF ULTRAFAST QUASI-PARTICLE RELAXATION TIME IN NORMAL METALS AS A FUNCTION OF TEMPERATURE USING TWO MODELS

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Abstract: Ultrafast quasi-particle relaxation dynamics for normal metal has been studied. Using the theoretical model developed by K. H. Ahn et. al. (2004) which is based on Two-Temperature model (TTM) and coupled Boltzmann equation, an evaluation of relaxation time τ for $LuAgCu_4$ metal has been performed. Our theoretically evaluated results are in good agreement with the experimental data and also with other theoretical workers.

Keywords: Ultrafast time-resolved optical spectroscopy, Femtosecond time- resolved pump-probe optical measurements, Time resolved terahertz spectroscopy, Picosecond time scale transient conductivity, Two- Temperature model (TTM), Coupled Boltzmann transport equation.

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INTRODUCTION

Ultrafast time-resolved optical spectroscopy has been used to reveal the nature of the quasi-particle relaxation dynamics in condensed matter systems. Femtosecond time-resolved pump-probe optical measurements have been carried out in normal metals [1,2], Conventional [3], high Tc superconductors [4] and charge-density wave solids [5]. Time-resolved terahertz spectroscopy has measured picoseconds time-scale transient conductivity in colossal magneto-resistance manganites [6]. These experiments show directly in the time domain how constituent degrees of freedom of materials interact with each other. This is important to understand the physics of governing the ground state and the low energy excited states of the materials. Understanding the fast dynamics of quasi-particles is also crucial for technological applications of these materials. These materials can be used to design very fast switching devices[7].

Recently femtosecond time-resolved pump-probe optical measurements on $LuAgCu_4$ and $YbAgCu_4$ were performed. There two materials are iso- structural with a negligible difference in lattice constant [8]. The differences of atomic masses are only 1/ between Lu and Yb. Their electronic structures are different due to f-shell occupancies in Lu and Yb. The closed shell 4f levels in Lu have no significant interaction Cu 3d and Ag 4d conduction electrons. In this way $LuAgCu_4$ is a normal metal [9]. On the other hand, the open shell 4f levels in Yb that is localized holes in the 4f [13] configuration have strong interaction with conduction electrons and make $YbAgCu_4$ a heavy fermion material. A heavy –fermion material is characterized by a large peak in the electron density of state (DOS) and the Fermi level E_F. The Sommerfeld coefficient γ is large[10,11]. The time-resolved optical experiments show very different relaxation dynamics in these two materials. $LuAgCu_4$ shows a relaxation time τ verses temperature (T) behavior similar to other normal metal such as Ag and Au[12]. In particular there is a little T dependence in τ at low temperatures. However, $YbAgCu_4$ shows approximately a100-fold increase in τ as T is decreased from Kondo temperature (T_K) down to 10K. The Kondo temperature for $YbAgCu_4$ is about 100K. This characterizes the width of the large DOS near E_F.

For normal metals and heavy-fermion materials ultrafast optical pump-probe spectroscopy was used. In this spectroscopy an ultrafast laser pulse initially excite the electron system and the probe pulse monitors the relaxation of the electron system. It measures the transient optical properties with sub picoseconds time resolution. Because of the diffusion of heat out of the illuminated (probed) region is much slower than the time scale of interest, the relaxation of the excited electrons is due to the thermalization among electrons and phonons within the illuminated area. For theoretical study of relaxation dynamics of photo-excited electrons in metals the most commonly used model is two temperature model (TTM). This model assumes

much faster relaxation within each constituent system (e.g electron system and phonon system) compared to relaxation between their constituent systems. In this approximation, the temperature of each system can be defined during relaxation and relaxation time τ between system1 and 2 and is determined by their specific heats C_1 and C_2 and the energy transfer rate per temperature difference g(T) [13]

$$\frac{1}{\tau} = g(T)(\frac{1}{C_1} + \frac{1}{C_2})$$

Kaganov, Lifshitz and Tanatarov [12] calculated the energy transfer rate g(T) between electrons (e) and phonons (p) in normal metals by solving coupled Boltzmann transport equation for electrons and phonons. They have taken thermal equilibrium distribution at different temperatures. Their result predict τ □ T at T> T_{Debye}/5 and τ □ T^{-3} for T < $_{TDebye}$/5 where T_{Debye} is the Debye temperature.

In this paper, we have evaluated the normalized excess electron energy as a function of time at high T (18meV=208 K) and low T(4meV=46K).We have also evaluated relaxation time as a function of temperature for $LuAgCu_4$ metal using TTM model and coupled Boltzmann equation. On comparing the evaluated results it appears that the results obtained from coupled Boltzmann equations are in good agreement with the experimental data [1,2].

MATERIALS AND METHODS

One considers the coupled Boltzmann equation for both electrons and phonons with electron-electron and electron-phonon scattering included[15]. Let $f_\varepsilon(t)$ and $f_\omega(t)$ be the electron and phonon distribution at time t. Let $D_e(\varepsilon)$ and $D_p(\omega)$ be the density of states (DOS) of electron and phonon respectively. Here ε and ω represent electron and phonon energies. The coupled Boltzmann equations are[16,17]

$$\frac{df_\varepsilon}{dt} = [\frac{df_\varepsilon}{dt}]_{ee} + [\frac{df_\varepsilon}{dt}]_{ep} \quad (1)$$

$$\frac{db_\omega}{dt} = [\frac{db_\omega}{dt}]_{ep} \quad (2)$$

The Collision integrals are

$$[\frac{df_\varepsilon}{dt}]_{ep} = \int d\omega K_{ep}\{ f_{\varepsilon+\omega}[(1-f_\varepsilon)(b_\omega+1) - f_\varepsilon(1-f_{\varepsilon+\omega})b_\omega\}$$
$$D_p(\omega)D_e(\varepsilon+\omega) + [\, f_{\varepsilon-\omega}(1-f_\varepsilon)b_\omega - f_\varepsilon(1-f_{\varepsilon-\omega})(b_\omega+1)]\; D_p(\omega)D_e(\varepsilon-\omega) \quad (3)$$

$$[\frac{db_\omega}{dt}]_{ep} = \int d\varepsilon K_{ep}[-b_\omega f_\varepsilon(1-f_{\varepsilon+\omega} + (b_\omega+1) f_{\varepsilon+\omega}(1-f_\varepsilon)]D_p(\varepsilon)D_e(\varepsilon+\omega) \quad (4)$$

$$[\frac{df_\varepsilon}{dt}]_{ee} = \int d\varepsilon' d\varepsilon'' \tfrac{1}{2} K_{ee}[-f_\varepsilon f_{\varepsilon'}(1-f_{\varepsilon''})(1-f_{\varepsilon+\varepsilon'-\varepsilon''})$$
$$+(1-f_\varepsilon)(1-f_{\varepsilon'}) f_{\varepsilon''} f_{\varepsilon+\varepsilon'-\varepsilon''}]D_e(\varepsilon')D_e(\varepsilon'')D_e(\varepsilon+\varepsilon'-\varepsilon'') \quad (5)$$

Here Kep and Kee are scattering matrix elements. The coupled Boltzmann equations are solved in two different ways.

(i) An condition at t=0 is set as a non thermal electron distribution excited by laser pulses, $f_\varepsilon(t=0) = f_{FD}(\varepsilon,T_i) + \Delta f_\varepsilon$ and thermal phonon distribution $b_\omega(t=0) = b_{BE}(\omega,T_i)$ where Ti and $f_{FD}(\varepsilon,T_i)$ and $b_{BE}(\omega,T_i)$ represent the initial temperature, Fermi Dirac and Bose-Einstein distribution function before the photo-excitation.

(ii) In the second method, one performs the linear stability analysis around the finial states $f_{FD}(\varepsilon,T_f)$ and $b_{BE}(\omega,T_f)$ by expanding the coupled Boltzmann equation. Equation (1) and (2) are linearly in $\delta f_\varepsilon(t)$ and $\delta b_\omega(t)$. We have

$$f_\varepsilon(t) = f_{FD}(\varepsilon,T_f) + \delta f_\varepsilon(t)$$
$$b_\omega(t) = b_{BE}(\omega,T_f) + \delta b_\omega(t) \quad (6)$$

Now taking $\delta f_\varepsilon(t) = V_\varepsilon^{\,e} \exp(-\frac{1}{\tau})$

$$\delta b_\omega(t) = V_\omega^{\,p} \exp(-\frac{1}{\tau}) \quad (7)$$

The linear differential equation has been cast into an eigen value problem of an (N+M)X (N+M) matrix. Now the solution gives $\frac{1}{\tau}$ and the normal modes of the relaxation. Two of the normal

modes have unphysical infinite relaxation time τ which originates from total energy and total electron number conservation. The rest of the modes represent all possible relaxation modes of the system. In general, the relaxation of the system can be represented as a linear combination of those modes. Now the energy transfer rate from electron system to phonon system at time t of specific mode with relaxation time τ is given by

$$dE_{\varepsilon,T}(t)/dt = [f(\varepsilon - E_F)(-\frac{1}{\tau})v^e{}_{\varepsilon} D_e(\varepsilon) d\varepsilon] e^{\frac{-1}{\tau}} \quad (8)$$

Now to identify e-p relaxation modes we have energy transfer strength

$$r_E(i) = \frac{1}{\tau(i)} \sum_{n=1}^{N} (\varepsilon_n - E_F) v^e{}_n D_{\varepsilon}(\varepsilon_n) \Delta\varepsilon \quad (9)$$

Where $\Delta\varepsilon$ is the energy step size, $v_n^{\ e}(i)$ is the normalized eigenvector for i th mode with proper overall sign. $r_E(i)$ Characterizes the effectiveness of i th mode for electron- phonon energy relaxation. The mode which has large $r_E(i)$ dominates in e-p relaxation and their eigenvector describe how the relaxation between electron and phonon system occurs. The mode with small $r_E(i)$ are either p-p or e-e relaxation modes and describe how the relaxation within each system happens.

For $LuAgCu_4$ we take[9,10]

$De=2.1eV^{-1}\ f.u^{-1}\ spm^{-1}$

Debye energy $\omega_D = 24meV$

$$D_P(\omega) = \frac{18\omega^2}{\omega^3{}_D}$$

$$\frac{K_{ee}}{K_{ep}} = 700$$

$$K_{ep} = 0.93eV/s$$

Here Kee and Kep are square of scattering matrix element. De and Dp are the final density of states for electron and phonon respectively. Kep is energy –independent constant

RESULTS AND DISCUSSION

In this paper, we have theoretically studied the ultrafast relaxation dynamics in normal metals. We have taken the theoretical formalism of K. H. Ahn et. al[18] in this study. We have evaluated normalized excess electron energy as a function of time for high temperature (T=18meV=208K) and low temperature (T=4meV=46K). The result is shown in table T1. Our theoretically evaluated results show that $\frac{\Delta E_e}{\Delta E_{e,\max}}$ as a function of time. The decrease is very fast for high T and slow for low T. In table T2, we have given the results of relaxation time as a function of time for high and low T. For both high and low T instantaneous relaxation time first decreases and then increases very fast and finally it attains a constant value. In table T3 , we have shown the relaxation time as a function of temperature T(K) foe normal metal $LuAgCu_4$. The evaluation has been performed by two models, one is TTM model (Two Temperature Model) and other is coupled Boltzmann transport equation. The evaluated results were compared with the experimental data. It was found that the obtained results are more near to coupled Boltzmann equation. In Table T4, we have shown the evaluated results of normalized $(\frac{df}{dt})$ and normalized $(-\frac{df_{FD}}{dt})$ for both high and low T. By comparing these two results, we examined whether the electron system approaches the final equilibrium state while maintaining the thermal distribution or not. In case of high T, the values of two normalized functions are close to energy 0.08eV showing that electron system has thermal equilibrium [19]. In Table T5, we have repeated the calculation of table T4 for low T. In this case, we noticed that at low T an electron system does not have thermal distribution. In Table T6, we have shown the evaluated results of relaxation time of $LuAgCu_4$ as a function of temperature for very large value of Kee as Kee=7000Kep. Calculations were performed again with two models TTM and coupled Boltzmann equation. We observed that an electron system has a thermal distribution in the whole T range. Now by increasing Kee to 7000Kep, the electron-electron relaxation becomes faster than the electron-phonon relaxation in the low T. At low T, TTM prediction give $\tau \propto T^{-3}$. Here Kee and Kep are square of scattering matrix element. The Boltzmann equation does not include direct phonon-phonon scattering and therefore the phonon distribution is non thermal except at t = ∞. One speculate very weak dependence of e-p thermalization dynamics on the phonon distribution in contrast to very strong dependence on thermal distribution. This originates from the characteristic of Bose versus Fermi statistics [20]. Our theoretically evaluated results are in good agreement with other theoretical workers [21-30].

CONCLUSION

From above evaluated results and investigation, we conclude that thermal electron distribution give rise to slow $\tau \square T^{-3}$

Relaxation behavior and the non-thermal electron distribution give faster and less T-dependence relaxation behavior at low T.

TableT1

An evaluated results of normalized excess electron energy versus time at high (T=18meV=208K) and low temperature (T=4meV=46K)

Here $\Delta E_e(t) = E_e(t) - E_e(t=\infty)$, $\Delta E_{e,\max} = \Delta E_e(t=0)$

t(ps)	$\frac{\Delta E_e}{\Delta E_{e,\max}}$	
	T=18meV=208K	T=4meV=46K
o.o	3.25	2.96
o.5	1.75	1.85
1,0	0.956	0.862
1.5	0.732	0.705
2.0	0.456	0.403
2.5	0.058	0.063
3.0	0.039	0.047
3.5	0.027	0.032
4.0	0.017	0.022
4.5	0.0072	0.0086
5.0	0.0037	0.0067
5.5	0.0024	0.0060
6.0	0.0016	0.0058

TableT2

An evaluated results of instantaneous relaxation time versus time at high (T=18meV=208K) and low temperature (T=4meV=46K)

t(ps)	T=18meV=208K	T=4meV=46K
o.o	4.55	4.72
o.5	3.27	4.38
1.0	1.45	3.59
1.5	1.48	2.73
2.0	1.62	2.10
2.5	2.56	1.87
3.0	3.58	2.54
3.5	6.29	4.68
4.0	7.56	5.86
4.5	8.48	6.29
5.0	7.52	7.36
5.5	7.39	7.47
6.0	7.26	7.50

TableT3

An evaluated result of instantaneous relaxation time τ as a function of temperature T for $LuAgCu_4$

Temperature (K)	Relaxation Time τ (ps)		
	TTM	Coupled Boltzmann	Expt. Value
50	1.02	0.37	0.28
100	0.92	0.39	0.31
120	0.87	0.40	0.35
140	0.77	0.43	0.38
150	0.42	0.45	0.43
170	0.27	0.53	0.46
200	0.38	0.59	0.48
220	0.43	0.67	0.52
240	0.52	0.60	0.54
250	0.50	0.58	0.56
270	0.48	0.54	0.58
300	0.53	0.56	0.60
350	0.59	0.62	0.59

TableT4

An evaluated results of normalized $\frac{df}{dt}$ and normalized (- $\frac{df_{FD}}{dt}$) for high T=18meV=208K as a function of ε (eV)

Energy ε (eV)	$\frac{\frac{df}{dt}}{(\frac{df}{dt})_{max}}$	$\frac{df_{FD}}{dt}$ (-)
-0.2	0.05	0.032
-0.15	0.12	0.107
-0.10	0.27	0.298
-0.05	0.38	0.375
0.0	0.69	0.706
0.05	0.13	0.186
0.08	-0.28	-0.305
0.10	-0.09	-0.183
0.15	-0.02	-0.056
0.20	-0.005	-0.012
0.30	0.027	0.014
0.40	0.095	0.016
0.50	0.128	0.152

Table T5

An evaluated results of normalized $\frac{df}{dt}$ and normalized (- $\frac{df_{FD}}{dt}$) for low T=4meV=46K as a function of ε (eV)

Energy ε (eV)	$\frac{\frac{df}{dt}}{(\frac{df}{dt})_{max}}$	$\frac{df_{FD}}{dt}$ (-)
-0.2	0.026	0.018
-0.15	0.037	0.022
-0.10	0.176	0.033
-0.05	0.292	0.047
0.0	0.538	0.182
0.05	0.312	0.254
0.08	0.243	0.162
0.10	0.425	0.195
0.15	0.082	0.225
0.20	0.035	0.246
0.30	0.064	0.278
0.40	0.095	0.305
0.50	0.106	0.325

Table T6

An evaluated results of relaxation time for $LuAgCu_4$ for very large $K_{ee} \geq 7000K_{ep}$ as a function of temperature T calculations are performed with Boltzmann Coupled Equation and TTM predictions.

T(K)	Relaxation Time τ (ps)	
	Boltzmann Coupled Equation	TTM predictions
10	132	167
50	78	120
80	56	88
100	32	52
120	18	49
150	9	27
170	6	18
200	17	24
220	24	31
240	35	42
250	47	50
300	56	58
350	67	60

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