Revealing the anomalous nonequilibrium carrier relaxation dynamics in C$_{60}$-related materials

Shota Ono*1), Hiroyuki Shima2), and Yasunori Toda3)

1)Department of Physics, Graduate School of Engineering, Yokohama National University, Yokohama 240-8501, Japan
e-mail: shota-o@ynu.ac.jp
2)Department of Environmental Sciences and Interdisciplinary Graduate School of Medicine and Engineering, University of Yamanashi, 4-4-37, Takeda, Kofu, Yamanashi 400-8510, Japan.
3)Division of Applied Physics, Faculty of Engineering, Hokkaido University, Sapporo, Hokkaido 060-8628, Japan

We theoretically demonstrate that the temperature ($T$)-dependence of photo-excited carrier relaxation time ($\tau$) in energy-gapped systems strongly depends on the rate of energy transfer from a longitudinal to transverse acoustic phonon mode. By considering the anharmonic coupling between them, we resolve an ambiguity in how to interpret the $T$-dependence of $\tau$ in alkali-doped C$_{60}$ superconductors.

Key words: fullerene, superconductivity, carrier relaxation, anharmonicity

1. INTRODUCTION

The rich and fascinating physics found in C$_{60}$-related materials has attracted great attention for the remarkable electronic properties. One of the most important properties in their compounds is energy-gap ($\Delta$) formation: superconducting gap at the transition temperature $T_c$=18K and 28K in potassium and rubidium doped C$_{60}$ crystals ($K_3C_{60}$ and $Rb_3C_{60}$), respectively [1,2]. More recently, it has been reported that the charge-density-wave gap opens at 60K in quasi-one-dimensional (1D) peanut-shaped C$_{60}$ polymers that is synthesized by electron-beam irradiation to C$_{60}$ films [3].

Femtosecond pump-probe spectroscopy has shown to be a useful tool to characterize ordered-phase in gapped systems. Up to now, almost all the families of gapped systems (Bardeen-Cooper-Schrieffer superconductors, high-$T_c$ cuprate superconductors, and charge/spin-density-wave materials) have shown the diverging behavior of photo-excited carrier relaxation time ($\tau$) near $T_c$ as temperature $T$ approaches $T_c$ from below [4,5,6,7,8,9,10,11]. Such the diverging behavior of $\tau$ has been well described by a model of Kabanov et al [4]. In contrast, it has been reported that the C$_{60}$-related materials show the lack of divergence near $T_c$; instead they show an increase in $\tau$ as $T$ is lowered from above to below $T_c$ [2,3]. The non-diverging behavior of $\tau$ cannot be explained by the model mentioned above. Therefore, it is necessary to develop a model that interprets both diverging and non-diverging behaviors of...
Recently, we have proposed a model that accounts for both behaviors of $\tau$ in the photo-excited carrier relaxation dynamics \[12\]. The basic assumption underlying our model is that the carrier relaxation is described by anharmonic phonon decay in which a longitudinal acoustic (LA) phonon mode decays into transverse acoustic (TA) modes. We have shown that by considering the LA-TA coupling effect the temperature dependence of $\tau$ is in agreement with experimental results of the quasi-1D peanut-shaped $C_{60}$ polymers \[3\] and high-$T_c$ superconductor $Bi_2Sr_2CaCu_2O_8+d$ \[10\]; the former shows the non-divergence of $\tau$ near $T_c$, while the latter shows the divergence.

In this paper, we demonstrate a specific application of our model to the unsolved problem, i.e., an interpretation of the relaxation dynamics of the alkali-doped $C_{60}$ crystals. We report that the non-diverging behavior of $\tau$ in these systems can be explained by the anharmonic phonon decay from LA to the TA phonon mode, which supports the validity of the model that we have proposed in Ref. \[12\].

2. MODEL

The assumptions used in our model are as follows: First, carriers produced by pump pulse absorption relax toward the single-particle state above the superconducting gap via the electron-phonon interaction \[13\]. Second, carriers above (below) the gap emit (absorb) a LA phonon with energy $\hbar \omega$ larger than $2\Delta$ (see Fig. 1), resulting in a quasi-equilibrium state between them \[14\]. Thus, the distribution of the photo-excited carriers and LA phonons with $\hbar \omega > 2\Delta$ is described by the Fermi and Bose distribution functions, respectively, with the time-dependent temperature $T'(t)$ where the magnitude of $T'(t=0)$ is determined by the initial pump pulse intensity \[4\]. On the other hand, the distribution of phonons with $\hbar \omega < 2\Delta$ is described by the Bose distribution function with $T(t) [ < T'(t)]$ because such the low-energy phonon cannot interact with carriers due to the energy-conservation law. Finally, our main assumption is that TA phonon modes can behave as the low-energy LA modes, irrespective to the TA mode energy. This is reasonable because there is no electron-TA phonon coupling within the deformation potential theory. This assumption means that the distribution of the TA mode is the same as that of the low-energy LA mode \[12\]. Thus, the Bose distribution functions for these phonon modes are given by

$$n(\omega) = \left[ \exp \left( \frac{\hbar \omega}{z} \right) - 1 \right]^{-1}$$  \hspace{1cm} (1)

where $z = k_BT'$ for LA modes with $\hbar \omega > 2\Delta$ and $z = k_BT$ for both LA modes with $\hbar \omega < 2\Delta$ and TA modes ($k_B$ is the Boltzmann constant.). Because of the

![Fig. 1: Schematic illustration of carrier relaxation dynamics across the energy gap. LA phonon mode emitted can be re-absorbed by a carrier below the gap or decay into TA modes.](image-url)
nonequilibrium phonon population generated by the pump pulse, the photo-excited carrier relaxation dynamics is governed by the anharmonic decay of the LA phonon with $\hbar \omega > 2\Delta$ into both TA and low-energy LA phonon modes, which is equivalent to the time-evolution between $T(t)$ and $T'(t)$. Substituting Eq. (1) into the Boltzmann transport equation that takes into account of the effects of three-phonon scattering, we obtain the relaxation time $\tau$ of the carriers [12]

$$\tau = \frac{C_{TA} + C_{LA}}{w_{CF}(T'') + w_{LL'}(T''')},$$  

where $C_{TA}$ ($C_{LA}$) is the heat capacity of TA (LA) phonons within the Debye approximation and $w_{CF}$ ($w_{LL}$) is the matrix element between the initial state with a LA phonon and final state with one or more TA (LA) phonons. The explicit expressions of $f(T)$ and $g(T)$ are determined by evaluating the collision terms of the Boltzmann equation and depend on the ambient temperature $T(0)$ [below we shall not distinguish $T$ and $T'(0)$], gap energy at absolute zero $\Delta(T = 0)$, and cutoff frequency of LA (TA) mode $\Omega_{LA}$ ($\Omega_{TA}$). For convenience, we define $p = w_{CF}/w_{LL}$. This is the anharmonic coupling strength between LA and TA mode.

If $p = 0$ and $C_{TA} = 0$ (i.e., there is no contribution from the TA phonon modes in the carrier relaxation), our model reduces to the model described in Ref. [4] that have explained the diverging behavior of $\tau$ at $T = T_c$.

3. RESULTS AND DISCUSSIONS

We turn next to a specific application of our model to the pump-probe data of the $C_{60}$-related materials [2]. In Figs 2(a) and 2(b), we plot the $T$-dependences of $\tau$ of K$_3$C$_{60}$ and Rb$_3$C$_{60}$, respectively, for different $p$'s using Eq. (2). As $p$ increases, $\tau$ decreases over the whole temperature range. As $T$ decreases, $\tau$ increases for all $p$'s that we have used here. It is clear that the theoretical curves are excellently agreement with the experimental data (crosses and solid circles [2]) when we set $2\Delta(0) = 3.5k_BT_c$, $\Omega_{LA} = 200K$, and $\Omega_{TA} = 35K$ (46K) for K$_3$C$_{60}$ (Rb$_3$C$_{60}$) [15]. These results unveil the carrier relaxation dynamics of the $C_{60}$ crystals.

Before closing, we mention briefly an open question.
For a complete understanding of the relaxation dynamics of various gapped systems, it is important to estimate the magnitude of $p$. It requires accurate first principles calculation of the matrix elements for the anharmonic interaction Hamiltonian. However, it is still a considerable task to obtain it correctly for complex systems. In fact, as far as we know, first principles calculations with regard to the matrix elements have been performed only on simple systems such as silicon and transition metals [16,17,18]. Thus, the theoretical prediction of the carrier relaxation dynamics of complex materials from a first-principles approach is a challenging task that we must tackle in the future.

4. CONCLUSIONS

We have theoretically shown that the non-diverging behavior of $\tau$ in alkali-doped $C_{60}$ materials can be explained by the LA-TA mode coupling that has not been recognized so far as an efficient relaxation channel in the photo-excited carrier relaxation dynamics. These results unveil an unconventional relaxation mechanism in $C_{60}$ materials, leading us closer to a unified interpretation of nonequilibrium carrier relaxation dynamics in gapped systems.

ACKNOWLEDGMENT

We thank K. Ohno for helpful discussions. This study was supported by a Grant-in-Aid for JSPS Fellows.

REFERENCES


(Received December 17, 2012; Accepted February 25, 2013)