Magnetic excitations investigated by ultrashort pulse excitation in high-T_c superconductors

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Abstract

We have investigated the relaxation of elementary excitations in underdoped Bi2212 by ultrafast Raman spectroscopy. In $\acute{x}\acute{y}$ symmetry, a broad feature has been observed around 2500 cm^{-1} in the Stokes side and assigned to B_{1g} two-magnon peak. In investigating the power dependence of the broad feature, we observed a super-linear behavior in B_{1g} symmetry, which is ascribed to the excess magnon population generated through the non-radiative relaxation across the CT gap. We observed a fast relaxation component around 0.4 ps and a long life time component greater than 20 ps for the B_{1g} two-magnon peak.

Key words: dynamics; Raman; magnon

1. Introduction

The interest in the dynamics of quasi-particle in high-T_c superconductors has recently attracted a lot of attention in terms of industrial applications[1]. Using ultra-fast Raman spectroscopy we can provide direct insights into the nature of fundamental carrier dynamics and interaction among the elementary excitations. Owing to the strong coupling of the CT excited state with magnons, we can expect to investigate dynamics of spin excitations by tracing the relaxation process starting from CT excitation. In addition, we analyze the results obtained from pump-probe time-resolved Raman measurements for the two-magnon peak. The non-radiative relaxation due to magnons strongly coupled to the CT excited state is suggested as a possible mechanism for the observed dynamics.

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2. Experiment

We used 100 femto second system with ultraviolet light [SHG (3.12 eV) of mode locked pulse laser] with a repetition rate of 82 MHz at 798 nm. The single crystals of $Bi_2Sr_2CaCu_2O_{8+\delta}$ which are used were prepared by the traveling solvent floating zone technique. The samples were mounted on the cold finger of a He flow cryostat with the temperature adjustable between 5 and 295 K.

3. Results and Discussion

Raman scattering spectra have been investigated in heavily underdoped sample with T_c=62 K. We observed a broad feature around 2000 cm^{-1} extending up to 3000 cm^{-1} in $\acute{x}\acute{y}$ configuration. This band is identified as the two-magnon continuum arising from AF ordered part, which is predominant in B_{1g} symmetry. In our previous report, we observed super-linear and sublinear power dependence for $\acute{x}\acute{y}$ and $\acute{x}\acute{x}$ polarization configurations, respectively [2]. In order to inves-

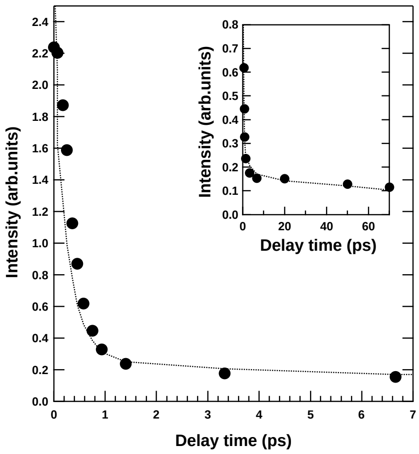

Fig. 1. Time response for the two-magnon peak in Bi2212 (T_C=62 K) with 100 femtosecond pulse excitation. The inset shows long time response.

tigate the origin of this non-linear power dependence, we performed pump and probe time-resolved Raman measurements. The Raman spectra were clearly different for different time delay measured from 0 up to 70 ps.

Figure 1 shows time-response for the two-magnon peak measured in $\acute{x}\acute{y}$ configuration at 5 K. We were able to fit the time response curve by exponential functions.

$$I(t) = K_1 e^{-t/\tau_1} + K_2 e^{-t/\tau_2} \quad (1)$$

The time constant was found to be 0.41 ps for τ_1, whereas τ_2 is shown to have a very long time response ($\tau_2 \approx 70$ ps). The long time response observed could be attributed to localized states very close to Fermi energy [3]. Thus we could decompose the relaxation process to both fast rate component and slow rate component and both are consistent with the measurement with ps pulse excitation [2]. We discuss the fast component in relation with the relaxation of elementary excitations which are coupled to CT gap. The photon energy of the pumping beam is much higher than that of charge transfer transition which is around 1.6 to 1.8 eV. There will be several mechanisms giving a time response. The relaxation time of 0.41 ps in Bi2212 is comparably fast, and radiative recombination can be excluded as an explanation for this recombination process. Since the energy quantum of magnon is several times larger than that of typical phonon mode in Bi2212, magnon emission might be the most important channel through which the CT excited state could be relaxed non-radiatively to the ground state. The carriers cooled down in the band by emitting phonons will further non-radiatively relax to the ground state by magnon-assisted interband transition as illustrated in Fig. 2.

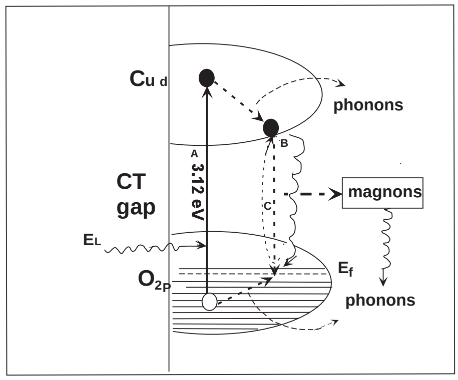

Fig. 2. Schematic energy diagram showing the relaxation process.

The excess magnon population will enhance Raman intensity and the time response is understood in terms of this non-radiative process.

The results of the pump and probe measurements show that the relaxation process including the backward process terminates within 0.41 ps.This corresponds to the life-time of the excited state of the charge-transfer excitation including magnon life time. In $\acute{x}\acute{x}$ configuration Raman intensity tends to increase with increasing time delay [2]. This is consistent with the sublinear power dependence and confirms that the origin of $\acute{x}\acute{x}$ broad feature is different from the origin of the two-magnon peak observed in $\acute{x}\acute{y}$ configuration. The origin of the $\acute{x}\acute{x}$ component has been a puzzle for a long time. However, there was no way to separate the origin of $\acute{x}\acute{x}$ and $\acute{x}\acute{y}$ only from spectral shape. By pulse excitation it becomes possible to recognize the diffrence of the origin of $\acute{x}\acute{x}$ and $\acute{x}\acute{y}$ broad feature.

In summary, we have investigated time-resolved Raman spectra of the two-magnon peak in underdoped Bi2212. We observed a fast relaxation component with a lifetime of 0.41 ps and a slow component with a lifetime of 70 ps. We have attributed the fast time component to the life time of non-equilibrium magnon. On the other hand, $\acute{x}\acute{x}$ configuration showed different time response. This clearly indicates that the origin of the peak observed in $\acute{x}\acute{x}$ is different from the two-magnon peak observed in B_{1g} symmetry.

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