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PUMP-PROBE SPECTROSCOPY WITH ENTANGLED PHOTONS

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A quasi-two-dimensional Hubbard Hamiltonian based on a four-site main cluster was established to simulate the $LaMnO_3$ system [1]. Theoretical investigation of the spin-orbital ordered $LaMnO_3$ using quantum light will be presented.
Keywords: $LaMnO_3$, phonon- coupled orbiton excitation, spectroscopy.

I. Introduction

Light-matter interactions can provoke optical excitations between the quantum states of the matter, which result into distinguishable features in infrared and Raman spectra. Analysis of the spectral features provides the information of the interactions in the many-body quantum states of the matter. In some solid matters such as strongly correlated electron systems, however lattice, charge, spin and orbital degree of freedom compete each other on similar energy scales to form the complicated low energy states [2], so numerous possibilities are allowed for optical excitations of microscopically different origins at similar energy scales, which causes ambiguity in the interpretation of the spectral features. The ambiguity can be reduced, if multiple light fields are employed as proposed in some non-linear optical techniques.

One challenge in condensed matter physics is the experimental confirmation of orbital wave or orbitons which is predicted to exist in an orbitally ordered state. Saitoh et al. have observed three peaks at 160,

144 and 126 meV in the Raman scattering of LaMnO3, and interpreted these as evidence of orbitons. But Grüninger et al found similar peaks in the optical conductivity of LMO and pointed out that the direct observation of orbitons in opical conductivity is prohibited by a selection rule. This suggested that the Raman peaks observed by Saitoh et al arise from multiphonons. Therefore, the quest for different techniques is still demanding.

Combinations of light fields with different frequencies allow the optical processes for different excitation-decay paths, which makes the intensity signals of the processes to appear at different places in the multidimensional frequency space composed of those light field frequencies. More information of the light-matter interaction process thus can be obtained from the multidimensional spectra[8]. Furthermore if those multiple light fields are entangled by their frequencies, fine tuning of the interference between the entangled light fields can make signal intensities of different excitation-decay paths to be different, which provides the chances to discriminate the light-matter interaction processes less ambiguously from the optical spectra. Entangled-light-field spectroscopic methods therefore may have advantage to resolve the low energy quantum states of the strongly correlated electron solid matter systems.

In this paper, we suggest that optical pump-probe method using polarization controlled entangled-photon pairs can be used to distinguish optical excitations of which resonant energy is very similar but the microscopic light-matter interaction process is quite different in a model system. For the demonstration, we numerically simulated the polarization dependence of the entangled-photon pump-probe response of a Mott insulator of which geometrical symmetry of the valence orbitals was broken.

II. Model and Method

A. Hamiltonian

We choose LaMnO3 as a model material system. This material system is well known for the spin and orbital ordering [1]. LaMnO3 has Mn^{3+} ions with electronic configuration $3d^4$ in high-spin state. Here one electron occupies one of the doubly degenerate valence orbitals. The orbitals show $3x^2-r^2=3y^2-r^2$ type ordering coupled to the JT lattice distortion and the spins show A-type ordering in the ground state. To mimic one lattice layer of the orbital and spin ordered LaMnO3 crystal, we built an ideal Mn-cluster composed of four Mn^{3+} ions [1].

B. Frequency-Dispersed Pump-Probe Signals

We propose a spectroscopy method based on the pump-probe process with two entangled light fields. Our method was inspired by Roslyak *et al.*'s work which demonstrated the pump-probe response of non-interacting molecules. In the proposed pump-probe spectroscopy, the pump and the probe light fields can be produced by the parametric down conversion (or parametric amplification) process in a nonlinear optical crystal such as $BaBO_2$. External excitation light with frequency ω_p and the wave-vector $\vec{k}_p$ illuminates the nonlinear crystal, then two entangled light fields (wave-vector $\vec{k}_1$, $\vec{k}_2$ and frequency ω_1, ω_2, respectively) were created. Due to the phase matching condition, the frequency (wave-vector) of the external excitation light field and the two entangled light fields should satisfy $\omega_p = \omega_1 + \omega_2$ ($\vec{k}_p = \vec{k}_1 + \vec{k}_2$).

We assumed that two entangled light fields interact with our model material system, i.e. the four-Mn-cluster. We considered ($\vec{k}_1$,ω_1) as the pump light field and ($\vec{k}_2$,ω_2) as the probe field. Then the pump-probe signal was generated by the transmittance change of ($\vec{k}_2$,ω_2) field caused by the interaction of our model system with ($\vec{k}_1$,ω_1) field. The arrival time of ($\vec{k}_2$,ω_2) field and that of ($\vec{k}_1$,ω_1) field on the sample were not well separated because of the same path length from the non-linear crystal to the sample in the proposed setup. We added a small modification, a half-wave plate (or periscope) into the original suggestion for the experimental setup by Roslyak *et al.* to control the polarization of the pump light field. Schematic diagram of the proposed setup was shown in Fig. 1.

The frequency-dispersed detection proposed here is expected to yield comparable but somewhat lower count rates[2]. The signal is given by

$$S(\Gamma, \omega, \delta t) = \frac{2}{\hbar}\Im\langle E_2^{\dagger}(\omega)P^{(3)}(\omega)\rangle,$$

where the third-order polarizability

$$P^{(3)}(\omega) = \int dt e^{i\omega t}P^{(3)}(t)$$

is induced by the interaction of the matter system with the light field. The set of control parameters Γ includes the pump pulse frequency ω_p and its bandwidth σ_p, the central frequencies of the downconverted beams ω_1 and ω_2, their entanglement time T specifying their bandwidths,

and the amplitude of the pump pulse α. δt is a variable delay. Expanding Eq. (1) to third order in matter-field interactions, we obtain the diagrams of Figure 2.

The pump light field ($\vec{k_1}$,ω_1) can generate many optical transition processes in the material, which can be described by the close-time-path-loop (CPTL) diagrams. Among them, eight CTPL diagrams are relevant to the pump-probe process in three-energy-level system according to the rotating wave approximation. The CTPL diagrams are called as the transition pathways. According to Roslyak *et al*., the pathways can be divided into two groups: two photon absorption (TPA) and stimulated Raman scattering (SRS). In the first group (TPA), the pathways showed that the system evolves from the ground state (g) to an excited state (e) by absorbing a light field, then to another higher energy excited state (f) by absorbing one more light field. Then it decays to a lower energy excited state (e) by emitting a light field, then finally to the ground state (g) by emitting one more light field. Full process can be represented shortly like $g \rightarrow e \rightarrow f \rightarrow e \rightarrow g$. All of these pathways involve the optical field correlation functions of the form $\langle a^\dagger a^\dagger a a \rangle$, where $a(a^\dagger)$ was the annihilation (creation) operator of the light fields. In the second group (SRS), the system undergoes the evolution from the ground state (g) to an excited state (e) by absorbing a light field, then come back to the ground state state (g) by emitting a light field, and the process is repeated once again. The pathway can be described shortly as $g \rightarrow e \rightarrow g \rightarrow e \rightarrow g$. All of the SRS pathways involve the optical field correlation functions of the form $\langle a^\dagger a a^\dagger a \rangle$.

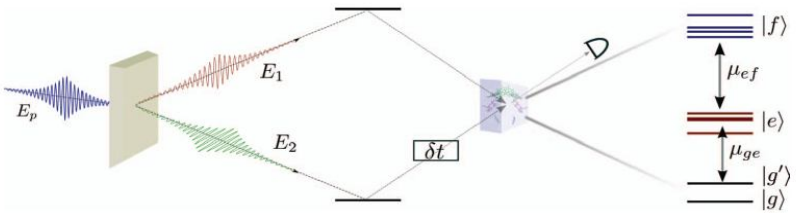

Fig. 1. Left: Setup for the pump-probe experiment with entangled photons. The two beams are generated by parametric down-conversion and interact with the sample modelled as a three-level system (right). The transmission of beam 2, which delayed by δt, is recorded. Right: Our three three-level system

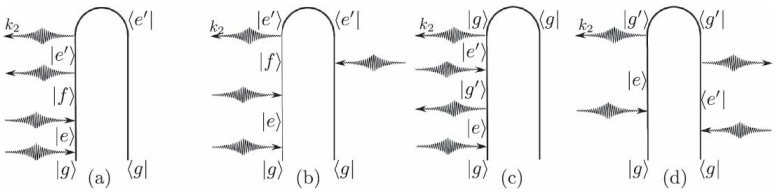

Fig. 2. Loop diagrams for the various pathways of the pump-probe signal. Unlabelled arrows represent the total field E_1+E_2. Diagrams (a) and (b) represent two photon absorption (TPA) pathways, and diagrams (c) and (d) are Raman pathways

IV. Conclusions

We have proposed an excitation-probe method using entangled photon pulses. Numerical simulation demonstrated that the two-photon absorption signal can be selectively filtered in the excitation-probe signal. By controlling the crystal thickness and the pump pulse frequency for the parametric downconversion, we can tune the bandwidth and the central frequency of the photon correlation function distribution used in the proposed method. We found that this tuning can be used to select specific transition peaks from the group of numerous stimulated Raman scattering peaks. We demonstrated that the proposed method can be used to examine the stimulated Raman scattering signal arising from orbital wave excitation in a model material, $LaMnO_3$.

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НАКАЧКА-ЗОНДАЖ СПЕКТРОСКОПИЯ СВЯЗАННЫХ ФОТОНОВ

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Квази-двумерный гамильтониан Хаббарда основанный на четырехмерном кластере был применен для вычисления системы $LaMnO_3$ [1]. Теоретическое исследование спин-орбитальногоLaMnO$_3$ с использованием квантового подхода будет представлено.
Ключевые слова: LaMnO3, фонон в сочетании, орбитон возбуждение, спектроскопия.