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Photoexcited carrier dynamics in potassium tantalate

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Abstract

Relaxation processes of photoexcited carriers have been studied in $KTaO_3$. The excitation above the absorption band edge generates a broad luminescence with a large Stokes shift, suggesting the formation of self-trapped excitons. The luminescence decay is found to be non-exponential, indicating the broad distribution of the carrier lifetime. The decay obeys power law after 10 ms, whose exponent becomes steeper as the temperature increases. The mean lifetime of the luminescence is as long as 10 ms at 4.6 K, and becomes faster at high temperatures. The temperature dependence of the lifetime should come from the thermally activated hopping motion of localized electrons and holes. Localized carriers may play an important role in the giant photoinduced dielectricity in $KTaO_3$.

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Keywords: Luminescence; Quantum paraelectrics; Decay; Self-trapping; Excitons

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1. Introduction

Quantum paraelectrics such as $SrTiO_3$ and $KTaO_3$ have recently attracted much attention since a drastic increase of the permittivity has been reported under band gap light irradiation [1–3]. The induced giant permittivity suggests the existence of microscopic fluctuations such as ferroelectric or metallic micro regions [3]. The theoretical studies of the excited states in the quantum paraelectrics have been made on the basis of the strong electron–phonon coupling and the anharmonic potential of the underlying lattice. They suggested that charge-transfer vibronic exitons [4], or superparaelectric polarons [5] should play an important role in the photoinduced states. However, the origin of this phenomenon has not yet been clarified, partially because of lack of the knowledge on relaxation processes.

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Therefore in order to understand the giant permittivity, we have made a series of luminescence experiments in $KTaO_3$ and discussed the nature and the dynamics of the photoexcited states using a simple model.

2. Experiments

We purchased single crystals of $KTaO_3$ from MTI Corporation. The crystals were grown by the slow-speed Flux method that can effectively avoid the incursion of oxygen vacancies. The sample was cooled in the He-bath cryostat with optical windows that enables us to measure the optical properties from 4.3 K to the room temperature. The second harmonic light (325 nm) of an optical parametric amplifier (10 Hz repetition rate, 2 ns pulse width) was used to excite the luminescence. The luminescence was dispersed by a 270 mm spectrometer and recorded by the liquid nitrogen cooled CCD camera. For the decay experiment, the luminescence was monochromated by the 250 mm spectrometer and detected by a photomultiplier tube (PM). The current output of the PM was recorded using an oscilloscope TSC5300 (Tecktronix Corp). We adjusted the load resistance from 50 Ω to 1 MΩ in order to obtain an appropriate time resolution and sensitivity. The timescale of our measurement extended from 10^{-5} to 0.1 s.

3. Result and discussion

Fig. 1 shows luminescence, absorption and excitation spectra in $KTaO_3$ at 4.3 K. It has been revealed from the two-photon absorption spectrum that $KTaO_3$ has an indirect band gap [6]. The indirect band-to-band absorption appears above 3.7 eV without any free exciton peaks. The absorption edge is found to obey the Urbach law, and the observed steepness parameter is around 0.6 at 5 K. The small value of σ indicates the stabilization of self-trapped excitrons (STEs) rather than free excitons, because the critical value to stabilize free or STEs is $\sigma = 0.94$ for the indirect band gap materials [7].

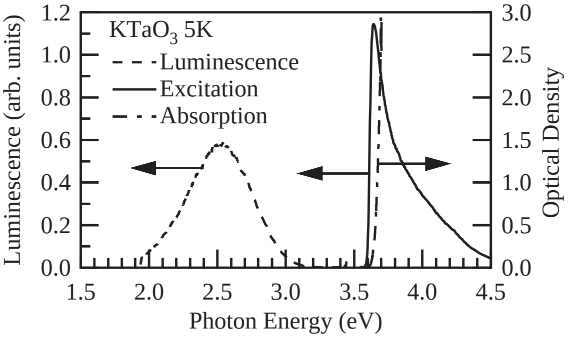

Fig. 1. Luminescence, excitation (left axis), and absorption (right axis) spectra of $KTaO_3$ at 5 K. The luminescence spectrum is obtained under excitation into 3.8 eV.

In $KTaO_3$, one can see a strong blue green luminescence band centered at 2.55 eV under band-to-band light excitation. The luminescence band has a broad bandwidth (0.8 eV) and a large Stokes-shift (1.05 eV), which strongly suggests a strong lattice relaxation of the excited state. Similar luminescence bands are observed in alkali halides [7] and in several oxides [8]. The origins have been attributed to the STEs or the oxygen vacancies that also induce a strongly relaxed exciton luminescence [9]. In $KTaO_3$, it has been revealed that the luminescence related to oxygen vacancies appears around 2.05 eV [9–11]. In Fig. 1, quite a weak 2.05 eV luminescence band can be seen in the low-energy tail of the 2.55 eV band. Therefore the 2.55 eV band is assigned not to the oxygen vacancies but to the intrinsic excited states. One possible candidate of the initial state of the 2.55 eV band may be STEs. The intensity of oxygen-vacancy-related luminescence band (2.05 eV) is quite weak compared with previous reports [10], reflecting that the crystal is well oxidized to contain little vacancies.

The excitation spectrum of the 2.55 eV band (detected at 2.48 eV) is also shown in Fig. 1. This demonstrates that the excitation edge is the same as the absorption edge. This correspondence supports the assignment that the 2.55 eV band should be intrinsic and comes from STEs. The gradual decrease in the high-energy side is usually connected to the non-radiative recombination centers on the surface of the sample [12]. In our case, we also imagine that the high-energy