Photocreation of a quantum domain and its detection by inelastic X-ray scattering and X-ray CARS

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Abstract. We briefly argue the concept of a quantum domain and propose its detection using X-ray inelastic scattering and X-ray coherent anti-Stokes Raman scattering (CARS). The quantum domain is defined as a spatial region of which the phase state is converted to a different one from that in the background. In a case where photoinduced phase transitions are allowed to exhibit, such a domain has a relatively low excitation energy and is expected to be detected experimentally. Especially the X-ray inelastic scattering and the CARS are attractive methods, since they can give information of both the momentum and the energy.

1. Introduction
The realization of material-phase switching is now a hot issue as often mentioned as "photoinduced phase transition." In particular, attention is paid to its ultrafast aspect, namely, electronic phase transition in a time scale of femto or subfemto seconds. The recent advance in synchrotron technology is now making it partially possible to detect such ultrafast phenomena. Needless to say, the advantage of using synchrotron radiation is in the full search of (q, ω) space. In particular, the q-scan gives a great merit, because a created domain of a photoinduced phase has a spatial internal structure and must be analyzed either in the real space or in the reciprocal space. Note that the real-space method is impossible here because of the translational invariance.

In a work performed a few years ago [1], we proposed the detection of a domain of an ionic phase in the neutral-phase background through stationary inelastic X-ray scattering. Following this work, we extended the theory to include the dissipation originating from scattering by phonons and found the effect of "quantum friction," that is, the deceleration of domain growth [2]. The current interest is in the detection of such a dissipating process by time-resolved experiment and we propose a detection based on coherent anti-Stokes Raman spectroscopy (CARS) in the X-ray region.

Below, we start our explanation from a model that describes the quantum domain, and briefly review the possible detection using the inelastic X-ray scattering. We also describe a proposed scheme of the X-ray CARS and give some calculated results for demonstration.

2. Model of quantum domain
First, we introduce our effective model hamiltonian, which is defined as

\[ H = H_D + H_{D-ph} + H_{ph}, \]
where the part for the bare domain in one dimension is expressed as

\[
H_D = -t_{\text{eff}} \sum_{jp} \{ (|j, p + 1 \rangle + |j - 1, p + 1 \rangle) \langle j, p | + H.c. \} \\
+ \sum_{jp} E(p) |j, p \rangle \langle j, p | ,
\]

the part of domain-phonon interaction as

\[
H_{D-ph} = g \sum_{i=j}^{j+p-1} (b_i + b_i^\dagger) |j, p \rangle \langle j, p | ,
\]

and the bare phonon part as

\[
H_{ph} = \omega \sum_j (b_j^\dagger b_j + \frac{1}{2}) .
\]

Regarding the details of the definitions, the readers are encouraged to see Ref. [2]. Instead, we describe our idea very briefly in the following. Namely, a one-dimensional domain as depicted in Fig. 1 is assumed to be photoexcited. Here, the state of the background phase is a type of electronic state, that is, a charge-density wave state, while that inside the domain is regarded as a Mott-insulator state. Next, we consider the eigenstates of the bare domain part, \( H_D \), for the special case of \( E(p) \equiv E_0 \). This case corresponds to the photoexcitation just at the phase boundary, which means that the formation energy is constant, \( E_0 \), irrespective of its size \( p \). In this case, the domain energy does not change even when it expands. Considering also the apparent fact that the domain has a translational symmetry, these basic properties lead to a picture that the two domain wall move freely to each other. As a result, we find a continuum band for the domain eigenstate as a function of its total momentum, \( K \), as shown in Fig. 2.

### 3. Inelastic X-ray scattering

In this section, we briefly discuss the inelastic X-ray scattering expected only for the \( H_D \) part. Since the part is already diagonalized for the case of \( E(p) = E_0 \), as \( \epsilon(k, K) = -4t_{\text{eff}} \cos(K/2) \cos(k) + E_0 \) with \( k \) being the internal momentum, we easily obtain expressions for the spectra. Namely, for \( K=\pi \), which corresponds to zero-momentum transfer, the spectrum becomes \( I(\Omega, \pi) = \delta(\omega - E_0) \). Meanwhile, it becomes a very broad spectrum expressed as
\[ I(\Omega, 0) = \frac{1}{\pi} \sqrt{1 - (\Omega - E_0)^2/(4t_{eff})^2}, \] as a normalized spectrum. Moreover, when \( E(p) \) is not constant, we have the results that were reported in Ref. [1], although an analytical treatment is impossible for this case. By the way, the validity of the present effective model was checked for the latter case, by the direct comparison between the result by this model and that by the original microscopic electronic hamiltonian. As the last remarks for this measurement itself, we point out the following two points. One is the large spectral width particularly for \( K=0 \). Considering that the \( t_{eff} \) is not so different from the bare electron transfer energy, the width as large as \( 8t_{eff} \) is rather outstanding, because even the bare electron has a band width of only \( 4t_{eff} \). Another point is the drastic band shrinking from \( K=\pi \) to 0, which suggests the experimental feasibility to detect such a feature.

4. X-ray CARS

In the previous section, we only considered the effect from \( H_D \). While such effect is very basic and predicts a coherent growth for the quantum domain, we also expect other effects that hinder such a growth. The domain-phonon interaction that was introduced in Eq. (3) plays such a role. Here, we assume an intramolecular type of vibrations, of which the boson operators are defined as \( b_j \) and \( b_j^\dagger \) at the \( j \)th site. After the photoexcitation by visible light, the initial total momentum is almost zero. However, in the course of the dynamics, the domain-phonon scattering occurs and hence the domain acquires a finite center-of-gravity momentum, \( K \), as depicted in Fig. 2. At the same time, such incoherence manifests itself as a deceleration in the domain growth [2]. What is important here is that such domain growth can never been observed by ordinary real-space imagines, since every process is translationally invariant except for special cases where some crystal imperfections or defects play some role. In this sense, it is required to to “visualize” such an evolution by using X-rays, not in the real space but in the \((q, \omega)\) space. From here on, we think about the case that two photons come in and other two photons come out, as illustrated in Figs. 3 and 4. To drive such a process, we utilize induced emission, which corresponds to the injection of \( k_2 \). Assuming the light pulse widths of \( \Delta \) and \( \Delta' \) and defining the delay time as \( \tau_d \), we find the scattering intensity as a function of \( K' \equiv (k_3) - (k_4) \) and \( \Omega' \equiv \omega_{k4} - \omega_{k3} \),

\[ S(\Omega', K') = \sum_{\nu} |D(K', p = 1)|_{ph}^2 \left| \frac{e^{i(\Omega' - \nu - H + E_{\nu})\Delta' - 1}}{\Omega' - H + E_{\nu}} \psi(\tau_d) \right|^2, \] (5)

Figure 3. Feynman diagram for the CARS. Rectangle shapes are assumed for the X-ray pulses.

Figure 4. Level scheme for the CARS. “x” is the direction of the one-dimensional axis.
where $\psi(\tau_d)$ is the state that evolved up to $t = \tau_d$, $|K',p = 1\rangle_D$ is the domain state with $K = 0$ and the size of $p$, and $|\nu\rangle_\nu$ and $E_\nu$ are the $\nu$th phonon state and its energy, respectively.

As the initial condition, we equal the $x$-component of $k_1$ and $k_2$, with $x$ being the one-dimensional direction, which corresponds to a preparation of the excited domain state with $K=0$ for the initial state. The probability of the evolved domain state as a function of $K$ and the energy then changes depending on the time, as a result of the domain-phonon scattering. Here, we calculate the time evolution exactly within a restricted Hilbert space of one domain and several phonons. In Fig. 5, we show a density map traced out for the phonon states, which is taken at $t=140$ in the unit of $\hbar$/eV $\sim 0.66$ fs. Note that $\Delta'$ is set at $2\Delta$ and $\Delta$ is 30 in the same unit. The other parameter values are the same as those in Ref. [2]. Although it is natural that the densities are distributed at the available domain eigenstates, the high densities around the band edges and, especially, at the crossing point should be paid attention, because they are related to the deceleration of the domain growth [2].

Next, we calculate the CARS intensities using the same final state, i.e., $\psi(\tau_d)$. In this study, we make one approximation, that is, the use of $H - H_{D-ph}$, instead of $H$ in the denominator of Eq. (5), neglecting the domain-phonon interaction in the short time of $\Delta'$. The obtained result is shown in Fig. 6. Although it is somewhat smeared out due to the finite observation time $\Delta'$, we recognize the same tendency as that in Fig. 5, which indicates the feasibility of this type measurement.

**Figure 5.** Probability density for the domain state with respect to $\epsilon$ and $K$ at $\tau_d=140$.

**Figure 6.** Two-dimensional map for the CARS at $\tau_d=140$.

### 5. Future Prospect

Through the detections discussed above, a new possibility will be opened in both the future X-ray spectroscopy and the research of the photoinduced phase transition. For those purposes, a new brilliant coherent light source is essential and its construction is highly waited to promote the science in these fields substantially.

### References

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[2] K. Iwano, Phys. Rev. B 84, 235139 (2011).