

# Quantum Many-Body Principles of Localized-State Ensemble Luminescence

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**Date:** 2026-02-06T17:42:47+00:00

## Abstract

Localized electron states induced by various types of disorder, including defects and impurities, commonly exist in solids. Their optical properties, especially their luminescence behavior, are of both fundamental scientific interest and technological relevance. However, a microscopic theory for luminescence from such localized-state ensembles (LSEs) has not yet been established. In this Letter, we seek to fill this gap by developing a quantum many-body (MB) luminescence theory that explicitly incorporates both electron-phonon (e-p) and electron-electron (e-e) interactions. Using the proposed MB-LSE framework, we quantitatively interpret a series of abnormal thermal behaviors, including redshift followed by blueshift of the emission peak, initial narrowing and subsequent broadening of the linewidth, reduction in emission intensity, and variations in the luminescence lifetime. The respective roles of electron-phonon and electron-electron interactions in variable-temperature LSE luminescence are thereby clarified. Moreover, within the MB-LSE theory, Varshni's empirical formula for the temperature dependence of the bandgap and the Huang-Rhys factor for e-p coupling are naturally derived and further discussed.

## Full Text

## Preamble

### Quantum Many-Body Principles of Localized-State Ensemble Luminescence

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## Abstract

Localized electron states induced by various disorders, including defects and impurities, commonly exist in solids. Their optical properties, particularly luminescence characteristics, hold both scientific and technological significance. However, a microscopic theory for such localized-state ensemble (LSE) luminescence has not yet been established. In this Letter, we attempt to fill this void by developing a quantum many-body (MB) luminescence theory that accounts for both electron-phonon (e-p) and electron-electron (e-e) interactions. Using the developed MB-LSE theory, anomalous thermal behaviors—including initial redshift followed by blueshift of the peak position, initial narrowing followed by broadening of the linewidth, intensity decline, and lifetime variation—can be quantitatively interpreted. The roles of electron-phonon and electron-electron interactions in variable-temperature LSE luminescence are thus elucidated. Moreover, within the framework of the MB-LSE theory, Varshni's empirical formula for bandgap temperature dependence and the Huang-Rhys factor for e-p coupling are further derived and discussed.

**Keywords:** Quantum many-body, localized-state ensemble luminescence, phonon, electron, temperature, thermal behaviors

## Introduction

Localized-state ensemble (LSE) luminescence is a ubiquitous physical phenomenon typically observed in luminescent solids containing certain disorders [?]. For example, anomalous temperature-dependent luminescence behaviors of localized states were discovered as early as the 1960s [?]. These include S-shaped shifts in luminescence peak position and non-monotonic variations in full width at half maximum (FWHM) [?]-[?]. On one hand, the existence of localized states can significantly benefit the performance of LEDs, lasers, and other optoelectronic devices [?]-[?]. On the other hand, the presence of localized states also poses challenges to the microscopic understanding and quantitative interpretation of LSE luminescence, despite the development of a quantitative model based on rate equations by one of the present authors and collaborators [?][?]. It is well recognized that LSE luminescence in solids represents an important photophysical phenomenon involving extremely complex many-body (MB) interactions. Unfortunately, to the best of our knowledge, a quantum mechanism-based MB microscopic theory has not yet been established for LSE luminescence in solids. Fortunately, the rapid development of quantum MB theory in recent years has brought new hope for solving this long-standing problem [?].

It is well known that emissive optical transitions of localized carriers are inextricably linked to their transport processes [?][?]. In this Letter, we develop a microscopic quantum theory for LSE luminescence. First, an effective Hamiltonian is proposed for an LSE system with Anderson localized states and MB interactions (including electron-electron (e-e) and electron-phonon (e-p) inter-

actions) [?]. Then, the time-resolved spontaneous emission probability of localized carriers in the LSE system is derived by determining the density matrix of the system. Furthermore, steady-state luminescence of the LSE system is obtained. Available experimental data—including temperature-dependent peak position, FWHM, integrated intensity, and lifetime of LSE luminescence—are quantitatively interpreted. Several puzzling mechanisms, including the roles of e-p coupling and e-e interactions in temperature-dependent LSE luminescence, are elucidated for the first time. Meanwhile, Varshni's empirical formula for bandgap shrinking and the Huang-Rhys factor for e-p coupling are derived from the developed MB-LSE luminescence theory. Therefore, the establishment of such an MB-LSE microscopic theory may provide novel and general understanding of the complex LSE luminescence in solids.

## Theoretical Model

In solids, the LSE system considered here may comprise many Anderson localized states and extended states. In such a system, quasi-particles such as electrons, holes, and excitons can move independently, recombine both radiatively and non-radiatively, and be scattered by phonons or captured and released by localized states, as schematically illustrated in Figure 1 [Figure 1: see original paper]. In path  $N + 1$ , a quasi-particle can enter and exit the LSE region. Although this particle may experience a complicated transport procedure in path  $N + 1$ , it does not produce light emission. In path  $N$ , however, light emission—i.e., photon generation—occurs.

**Figure 1.** A schematic picture illustrating transport and luminescence of quasi-particles from the launch source to the receiving terminal via an LSE system in the phonon field. Launching quasi-particles may have a certain probability of radiative recombination to produce luminescence in the LSE region. During transport and luminescence processes, the quasi-particles may be scattered by phonons. The chemical potential energies of the launch system and receiving system are  $\mu_L$ ,  $\mu_R$ , respectively, while the tunneling level of the LSE is  $E_T(\mu)$ .

As schematically shown in Figure 1, three processes that quasi-particles launched from the launch system ( $\mu_L$ ) may experience include: (1) Direct arrival at the receiving system ( $\mu_R$ ). (2) Scattering by optical phonons ( $ph(O)$ ) and acoustic phonons ( $ph(A)$ ) at levels  $E_L^{ph(O)}(\mu)$ ,  $E_L^{ph(A)}(\mu)$ , respectively, followed by radiative or non-radiative recombination and/or continuous transport in the LSE region. (3) Radiative recombination ( $R$ ) in the LSE region. Disregarding quasi-particle spin and assuming that only one quasi-particle is allowed to pass through an energy level channel, the energy levels for phonon scatterings are assumed to be far away from the tunneling energy level  $E_T(\mu)$  of the LSE system [?]. It should be noted that radiative recombination and phonon scattering of quasi-particles can occur at any point within the central LSE system. Of course, direct transport between the launch and receiving ends can also take place. Under such circumstances, an effective Hamiltonian describing transport, phonon scatterings, particle-particle interactions, and luminescence may be written as:

$$\begin{aligned}
 \mathcal{H}_{eff} &= \mathcal{H}_0 + \mathcal{H}_{e-p} + \mathcal{H}_{e-e} \\
 \mathcal{H}_0 &= \sum_i E_i \hat{c}_i^\dagger \hat{c}_i + E_L \hat{c}_L^\dagger \hat{c}_L + E_R \hat{c}_R^\dagger \hat{c}_R + \sum_{k,\alpha} E_L^{ph(\alpha)}(k) \hat{a}_{L,k,\alpha}^\dagger \hat{a}_{L,k,\alpha} \\
 &\quad + \sum_{k,\alpha} E_L^{ph(\alpha)}(k) \hat{a}_{L,k,\alpha}^\dagger \hat{a}_{L,k,\alpha} \\
 \mathcal{H}_{e-p} &= \sum_{k,\alpha} \Omega_L^{ph(\alpha)}(k) \left( \hat{c}_L^\dagger \hat{c}_L \hat{a}_{L,k,\alpha}^\dagger + \hat{c}_L^\dagger \hat{c}_L \hat{a}_{L,k,\alpha} \right) \\
 &\quad + \sum_{k,\alpha} \Omega_L^{ph(\alpha)}(k) \left( \hat{c}_L^\dagger \hat{c}_L \hat{a}_{L,k,\alpha}^\dagger + \hat{c}_L^\dagger \hat{c}_L \hat{a}_{L,k,\alpha} \right) \\
 \mathcal{H}_{e-e} &= \sum_{i,j} \Omega_{ij} \hat{c}_i^\dagger \hat{c}_j^\dagger \hat{c}_j \hat{c}_i + \sum_{i,j,k} \Omega_{ijk} \hat{c}_i^\dagger \hat{c}_j^\dagger \hat{c}_k^\dagger \hat{c}_k \hat{c}_j \hat{c}_i
 \end{aligned}$$

Detailed definitions of various quantities in the sub-Hamiltonians can be found in the Supplementary Information (SI) document. Referring to the outstanding work by Gurvitz and Prager [?], two interrelated differential equations in terms of density matrices for the probability of a particle being undetected ( $\sigma_{uu}$ ) and detected ( $\sigma_{dd}$ ) in the transport space can be eventually obtained:

$$\begin{aligned}
 \frac{\partial \sigma_{uu}}{\partial t} &= -\Gamma \sigma_{uu} + \Gamma \sigma_{dd} + \Gamma_{ph(O)} \sigma_{uu} + \Gamma_{ph(A)} \sigma_{uu} \\
 \frac{\partial \sigma_{dd}}{\partial t} &= \Gamma \sigma_{uu} - \Gamma \sigma_{dd} - \Gamma_{ph(O)} \sigma_{dd} - \Gamma_{ph(A)} \sigma_{dd}
 \end{aligned}$$

In Eq. (2),  $\Gamma$  gives the average width of the level due to interparticle interactions, where  $\Gamma = 2\pi\rho|\Omega|^2$  is the coupling strength, and  $\rho$  denotes the effective occupation density of states (DOS) for the corresponding particle. Derivation of Eq. (2) can be found in the SI document. By solving Eq. (2) with the initial conditions of  $\sigma_{uu}(0) = 1$  and  $\sigma_{dd}(0) = 0$ , one obtains:

$$\sigma_{uu}(\mu, T, t) = e^{-Xt}, \quad \sigma_{dd}(\mu, T, t) = \frac{\Gamma}{X} (1 - e^{-Xt})$$

where  $X = \Gamma + \Gamma_{ph(O)} + \Gamma_{ph(A)} + \Gamma_{re} + \Gamma_{re-exc}$ .

It should be noted that for any quasi-particles, if they are successfully transported from the launch system to the receiving end, they would be unable to emit photons, regardless of what complicated processes they experienced. For a given quasi-particle, its path consists of  $N + 1$  events in the LSE region, although it may have undergone multiple transport events between lattice points before entering the LSE zone. Now let us consider a quasi-particle undergoing transport (*tr*), entering the LSE space, and subsequently experiencing phonon scattering (*sc*), radiative recombination (*re*), and re-excitation (*re - exc*). If the total duration of this process coincides precisely with the time required for

the LSE luminescence intensity to decay to its  $1/e$  value, then the occurring probability (in natural units  $\hbar = 1$ ) of this event may be written as:

$$P(\mu, T, t_{tr}, t_{sc}, t_{re}, t_{re-exc}) \propto \sigma_{uu}^{(tr)}(\mu, T, t_{tr}) \sigma_{dd}^{(sc)}(\mu, T, t_{sc}) \sigma_{dd}^{(re)}(\mu, T, t_{re}) \sigma_{dd}^{(re-exc)}(\mu, T, t_{re-exc})$$

where  $t_{tr}$ ,  $t_{sc}$ ,  $t_{re}$ , and  $t_{re-exc}$  respectively represent the number of occurrences and the time at which their respective physical processes take place, and  $t_{tr} + t_{sc} + t_{re} + t_{re-exc} = t_{total}$  is the total time. If there are  $N$  localized states whose transport and recombination can occur within unit time and the DOS distribution of the localized-state ensemble is  $\rho(\mu)$ , then an approximate expression for time-resolved LSE luminescence intensity may be expressed as:

$$I_{tr} \propto PN \int \rho(\mu) P(\mu, T, t) d\mu = PN \int \rho(\mu) e^{-X(\mu, T)t} d\mu$$

where  $P$  represents the radiative recombination probability. Furthermore, the integrated intensity of steady-state LSE luminescence may be calculated with:

$$I_{ss} \propto PN \int \rho(\mu) \int_0^\infty e^{-X(\mu, T)t} dt d\mu = \bar{P}N \int \frac{\rho(\mu)}{X(\mu, T)} d\mu$$

where  $\bar{P}$  denotes the average radiative recombination probability of all quasi-particles in the LSE ensemble, which arises from re-excitation processes. Here,  $\bar{P} = \left( \frac{\Gamma_{re}}{\Gamma_{re} + \Gamma_{re-exc}} \right)$  represents the average number of re-excitation events. In addition, Gibbs free energy loss (GEL) of quasi-particles via emission of phonons should be considered. Therefore, photon energy of radiative recombination may be corrected to:

$$E = \mu - \hbar\omega_{ph(A)} \cdot \Gamma_{ph(A)} \cdot \bar{t}_{sc}$$

where  $\hbar\omega_{ph(A)}$  is the average time since the last scattering event occurred, and its value should be on the same order of magnitude as the luminescence lifetime. If the quasi-particle thermal equilibrium state is neglected, an analytical expression for the peak position can be derived (by referring to SI 2).

## Theoretical Photoluminescence (PL) Spectra and Comparison with Experimental Spectra

To test the applicability of the above-derived microscopic theory for LSE luminescence, we compare theoretical PL spectra with available experimental spectra from literature. Figure 2 Figure 2: see original paper shows the PL spectral data (solid squares) of InGaN quantum wells by Cho et al. [?]. Solid lines represent the theoretical spectra calculated with the new theory (denoted as MB-LSE

model hereafter). Agreement between theory and experiment is satisfactory for such a complicated solid system. It is also found that the PL line profiles become relatively symmetric with increasing temperature, reflecting thermal redistribution of quasi-particles within localized states [?]. Temperature-dependent peak positions, FWHM, integrated intensities, and decay times of both theoretical (solid lines) and experimental data (solid squares) are shown in Figure 2(b). The S-shaped temperature dependence of peak positions and distinct nonmonotonic variation of PL decay times can be well reproduced by the MB-LSE model, while some deviation exists for integrated intensities at low temperatures.

Figure 2(c) depicts calculated time-resolved PL spectra at different temperatures. At low temperatures, the theoretical PL spectra exhibit distinctively asymmetric line shapes with a deep high-energy side. As temperature increases, however, the line shapes tend to become symmetric predominantly due to thermal redistribution of localized quasi-particles within the localized states. The roles of e-p and e-e interactions in temperature-dependent PL behaviors are discussed below.

**Figure 2.** (a) Theoretical PL spectra (solid lines) with the newly developed MB-LSE model and experimental data (gray squares) of InGaN quantum wells by Cho et al. [?] for different temperatures. (b) Temperature-dependent peak positions, FWHM, integrated intensities, and decay times (from top to bottom). Solid squares represent experimental data by Cho et al., and solid lines stand for theoretical results. (c) Calculated time-resolved PL spectra at different temperatures of 10, 100, 200, and 300 K with the newly developed MB theory. For relevant calculation parameters, refer to SI 3.

First, the GEL effect was theoretically examined. If the interactions between quasi-particles and phonons were independent of their free energy, only the temperature dependence of peak position in the medium-high temperature range would be affected—i.e., the peak position would blueshift at about 50 K and tend to saturate at high temperatures, as shown by the red line in Figure 3 Figure 3: see original paper. Theoretical dependence without considering the GEL effect deviates significantly from experimental data (solid squares) by Cho et al. in the temperature range of 50-300 K. Relevant calculations were also performed for other materials, such as GaAs-based materials, with results shown in SI 4. If e-p interactions were not considered, as shown by the blue line in Figure 3, for peak position and FWHM, e-p interactions promote blueshift and broadening, respectively, by influencing wave function evolution. For integrated intensity and luminescence lifetime, both theoretical integrated intensity and lifetime rapidly increase with temperature in the absence of e-p interactions, which contrasts with experimental dependence (solid squares). In the low temperature range (< 50 K), the experimental lifetime growth may be interpreted as weaker e-p interactions. Additional calculations are provided in SI 5. Therefore, e-p interactions play a significant role in determining thermal behaviors of solid-state LSE luminescence.

**Figure 3.** Calculated temperature dependence of PL spectral parameters for

several cases, i.e., with and without e-p and e-e interactions. (a) Peak position, (b) FWHM, (c) Integrated intensity, and (d) Decay time. Solid squares represent experimental data from Cho et al. [?].

Now it may be helpful to offer a possible microscopic explanation for the famous Varshni' s empirical formula describing redshift temperature dependence of bandgaps in many materials [?]:

$$E_{gap}(T) = E(0) - \frac{\gamma T^2}{\theta + T}$$

By extremizing the Gaussian-type DOS distribution to a single energy level distribution and making a second-order Taylor expansion of Eq. (7), we may obtain the following approximate expressions:

$$\bar{t}_{sc} \propto \frac{1}{\Gamma_{ph(A)}}, \quad \gamma \propto \frac{2\pi\rho_{ph}|\Omega_{ph}(E_T)|^2}{\hbar\omega_{ph(A)}}$$

where  $k_B$  is a dimensionless factor and  $\theta \propto \frac{\hbar\omega_{ph(A)}}{k_B}$ . Here  $\hbar\omega_{ph(A)}$  is related to the acoustic phonon frequency (i.e., Debye cut-off frequency). Therefore, e-p interactions are the major mechanism causing Varshni' s redshift temperature dependence of the fundamental bandgap in semiconducting and insulating materials.

Regarding e-p interactions, the well-known Huang-Rhys factor ( $S$ ) [?] should be discussed because it is a pivotal parameter characterizing e-p coupling strength and thus determining some optical properties of solids [?]. On the premise that Gibbs free energy loss associated with the quasi-particle is the sole contributor to modification of lattice elasticity, we derive:

$$S = f \cdot n_{ph(A)}(\omega)$$

where  $n_{ph(A)}(\omega)$  denotes the average number of phonons with angular frequency  $\omega$ . If we set  $f = 2S^2$ , then the shift in peak position of LSE luminescence caused by GEL is:

$$\Delta E_{GEL} = 2S^2\hbar\omega_{ph(A)} = 2S^2\hbar\omega_{ph(A)}n_{ph(A)}$$

This is O' Donnell-Chen' s empirical formula for redshift temperature dependence of bandgap [?], which is well consistent with the e-p coupling term. The Huang-Rhys factor is thus a comprehensive function of temperature, phonon energy, and phonon scattering time. In addition, when  $\Gamma_{ph(A)} \rightarrow 0$ , the S-shaped temperature dependence of peak position of LSE luminescence can diminish and tend to a normal Varshni-type temperature dependence, as shown in Figure 4 [Figure 4: see original paper]. In fact, from Eq. (6), a maximum redshift

amount can be shown to be less than  $\Delta E_{max} < \frac{\Gamma_{ph(A)}}{\Gamma_{ph(A)} + \Gamma_{re}} \cdot \hbar\omega_{ph(A)} \cdot \bar{t}_{sc}$ . Other parameter definitions can be found in SI 1.

**Figure 4.** With decreasing  $\Gamma_{ph(A)}$  from  $\Gamma_{ph(A)}^{max}$  to zero, peak-position redshift at about 50 K gradually diminishes, resulting in a normal Varshni-type temperature dependence.

Besides e-p coupling, e-e interactions represent another important factor affecting LSE luminescence properties. They can also induce variations in energy level width through the thermal distribution of quasi-particles with disparate energies and modulation of coupling strengths. It is revealed that variations in e-e coupling strength significantly influence the thermal properties of LSE luminescence. When variations in e-e coupling strength were not considered, temperature dependence of all four theoretical spectral parameters (purple curves in Figure 3)—including peak position, FWHM, integrated intensity, and lifetime—substantially deviated from experimental trends (solid squares in Figure 3). Additionally, as detailed in Section SI 6, Coulomb interactions between electrons also influence injection and extraction of electrons in localized states. Finally, upon considering combined effects of e-p and e-e interactions, temperature dependence of luminescence intensity of a two-level system can be approximated as  $\zeta(T)e^{-E_a/k_B T}$  and  $\zeta(T)e^{-T/T_0}$  at low and high temperatures, respectively. Here  $\zeta(T) = \left(\frac{\Gamma_{re}}{\Gamma_{re} + \Gamma_{re-exc}}\right)^{N_{re-exc}}$  and  $E_a \approx \hbar\omega_{ph(A)} \cdot \frac{\Gamma_{ph(A)}}{\Gamma_{ph(A)} + \Gamma_{re}}$  at low temperatures and  $E_a \approx \Gamma_{re} + \Gamma_{re-exc}$  at high temperatures, respectively. Then the LSE luminescence intensity can be approximated as the famous Arrhenius form at low temperatures, and it can also be approximated as the laser temperature characteristic formula with a distinct re-excitation process [?][?].

## Conclusions

In summary, a microscopic many-body quantum theory is developed for localized-state ensemble luminescence in solids. In this theory, an effective Hamiltonian considering MB interactions is proposed and solved. The newly developed MB luminescence model is then applied to quantitatively interpret available temperature-dependent steady-state and time-resolved PL spectra of the InGaN/GaN multi-quantum-well system by Cho et al. Temperature dependence of several key spectral parameters—including peak position, FWHM, integrated intensity, and lifetime—can be well elucidated. For the unusual blueshift of the luminescence peak frequently observed, theoretical analysis indicates that variation in luminescence probability caused by e-p interactions plays a key role, whereas redshift of peak position in the high-temperature region usually originates from Gibbs free energy loss. For the FWHM of LSE luminescence, phonon scattering causes broadening. Meanwhile, phonon scattering plays a major role in integrated intensity attenuation and lifetime decline with temperature, especially at high temperatures. It is also unraveled that e-e interactions, particularly their variations with temperature, constitute an important factor influencing LSE luminescence characteristics at low

temperatures—for instance, resulting in redshift of peak position, narrower FWHM, and increased integrated intensity and decay time. The MB-LSE theory developed in the present study can be used to derive expressions for several well-known formulas and parameters, such as Varshni's empirical formula and the Huang-Rhys factor.

**Acknowledgements:** The work was financially supported by the National Natural Science Foundation of China (No. 12074324). The authors wish to thank Dr. Debao Zhang, Dr. Ji Zhou, Mr. Wanggui Ye, Mr. Xuguang Cao, Mr. Sicheng Liu, Mr. Ke Yu, and Prof. Jiqiang Ning for their support and important contributions to the work.

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