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Polariton condensates at room temperature

Condensats de polaritons à température ambiante

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ABSTRACT

We review the recent developments of the polariton physics in microcavities featuring the exciton–photon strong coupling at room temperature, and leading to the achievement of room-temperature polariton condensates. Such cavities embed active layers with robust excitons that present a large binding energy and a large oscillator strength, i.e. wide bandgap inorganic or organic semiconductors, or organic molecules. These various systems are compared, in terms of figures of merit and of common features related to their strong oscillator strength. The various demonstrations of polariton laser are compared, as well as their condensation phase diagrams. The room-temperature operation indeed allows a detailed investigation of the thermodynamic and out-of-equilibrium regimes of the condensation process. The crucial role of the spatial dynamics of the condensate formation is discussed, as well as the debated issue of the mechanism of stimulated relaxation from the reservoir to the condensate under non-resonant excitation. Finally the prospects of polariton devices are presented.

RÉSUMÉ

Cette article de revue est consacré aux récents développements de la physique des polaritons dans les microcavités présentant le couplage fort exciton–photon à température ambiante, aboutissant à la réalisation de condensats de polaritons à température ambiante. De telles cavités contiennent des couches actives spécifiques, dont les excitons présentent une grande énergie de liaison et une grande force d'oscillateur, i.e. des semiconducteurs à grand gap ou organiques, ou des molécules organiques. Les différents systèmes étudiés à ce jour sont comparés, sur la base de leurs figures de mérites et de leurs propriétés communes liées à leur grande force d'oscillateur. Cette comparaison s'étend ensuite aux différentes démonstrations de laser à polariton, et aux diagrammes des phases correspondant. Le fonctionnement à température ambiante permet en effet une étude détaillée des régimes thermodynamique vs hors d'équilibre du processus de condensation. Le rôle crucial de la dynamique spatiale de formation du condensat est aussi abordé, ainsi que la question encore débattue du mécanisme de relaxation stimulée depuis le

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réservoir jusqu'au condensat dans le cas de l'excitation non résonante. Enfin, les enjeux des dispositifs polaritoniques sont présentés.

In the past decade, the semiconductor microcavities have been widely used for both the fundamental studies of the polariton collective states such as condensates and quantum fluids, and the demonstrations of original quantum optoelectronic devices: polariton lasers, switches, transistors. For both topics, the prospect of an operation at room temperature has raised a strong interest and led to the development of specific microcavities based on new active layers. The key requirements for strong coupling up to room temperature are large binding energy and large oscillator strength of the excitons; a broad spectrum of active layers is presently being explored among inorganic semiconductors (mainly GaN, ZnO, ZnSe, CuCl, CuBr) and organic molecules (dyes, J-aggregates, perovskites). Their integration into optical cavities jointly requires a strong technological effort, so to reach quality factors (Q) typically larger than 1000. Some of those room-temperature microcavities are now mature enough to allow in-depth fundamental studies of the polariton condensates, their formation, propagation and control, as well as examples of polariton devices. We give here an overview of the recent demonstrations achieved with such microcavities, and emphasize their specificities compared to GaAs and CdTe-based microcavities.

1. Robust excitons for room-temperature polaritons

In the choice of the appropriate active layer, the main criterion for a room-temperature operation of a microcavity is the following: the exciton binding energy and the polariton Rabi splitting should be both larger than the thermal energy at 300 K ($k_B T = 26$ meV). The search for the strong exciton–photon coupling at room temperature therefore started with organic molecules in 1998 [1] and wide bandgap semiconductors, especially GaN in 2003 [2–4]. ZnO [5–9] and CuCl [10] rapidly emerged as alternative candidates for strong coupling, up to $T = 550$ K in ZnO [11,12], as well as ZnSe [13–16] and CuBr [17]. Organic active layers include perovskites [18,19], J-aggregates [1,20], anthracene [21], fluorene (TDAF) [22], conjugated polymers [23]. As detailed in Table 1, all those materials exhibit an exciton binding energy larger than 26 meV, as well as a longitudinal-transverse splitting larger than a few meV (compared to 80 μeV for GaAs), leading to large Rabi splittings. It should be noticed that bulk active layers and quantum wells are both deeply investigated as active layers.

Once the active layer is inserted inside a microcavity, the Rabi splitting deduced from the mode anticrossing assesses the strong coupling regime between excitons and photons (see Table 1), as probed by reflectivity or transmission measurements. The main impediment towards polariton luminescence, polariton lasing or condensation is the improvement of the microcavity quality factor on the one hand, and the radiative efficiency of free excitons at 300 K on the other hand. For each material, this requires an intense technological effort in order to maintain an excellent crystalline quality of the active layer while improving the cavity features. For GaN-based microcavities, this led to the development of crack-free lattice-matched nitride distributed Bragg reflectors (DBRs) [24], and more recently of crack-free nitride DBRs on patterned silicon substrates [25]. For other materials, a complex combination of nitride DBRs and dielectric DBRs is often implemented, while preserving the quality of the active layer [26,27].

While most works have been reported with planar microcavities, alternative optical resonators are also explored. The possibility of a self-organized growth of GaN and ZnO into hexagonal nanowires [28–30] and microwires [11,12,31–34] has led to interesting resonators whose crystalline quality is improved compared to planar structures. The exciton then couples with either longitudinal Fabry–Perot modes in cleaved nanowires (like in a finite length monomode optical fiber), or whispering gallery modes in micron-thick wires. The strong coupling has been demonstrated, with larger Rabi splittings than in planar microcavities, due to the optimal overlap between the photon mode and the free exciton (see Table 1). Moreover, this offers the possibility to explore 1D polaritonic systems without requiring the implementation of the complex lithography and etching processes presently used to obtain 1D polaritons in GaAs wire-shaped cavities [35].

As discussed in the following sections, the achievement of a good photon confinement in the resonators appears as a requirement for the further demonstrations of polariton lasers. The minimum quality factor of the photonic modes is of the order of 1000 in the case of GaN and ZnO active layers, and diminishes to 500 for CuBr and 200 for anthracene, both with larger exciton binding energies.

2. Impact of the large oscillator strength on the polariton eigenmodes

The impact of the large oscillator strength of the excitonic transitions has been studied in details in the case of ZnO. In early works on ZnO nanowires [28,36], the deviation between the Fabry–Perot free spectral range far and near the excitonic transitions is discussed as a first signature of the strong coupling between excitons and confined photons. Indeed the refractive index of ZnO presents pronounced resonances corresponding to the exciton states as shown in Fig. 1a, so that the textbook resonance condition for the Pth Fabry–Perot resonance $n_{ZnO}(\lambda) d_{ZnO} = P\lambda/2$ is fulfilled for a few values of P. Let us compare the cases of two planar microcavities embedding a bulk GaAs and a bulk ZnO active layer. In the GaAs microcavity [37], the cavity features are understood through the introduction of the GaAs refractive index (real part) in the

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