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Room Temperature Superfluorescence from an Electron-Hole Liquid

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Superfluorescence, a coherent burst of light from an excited ensemble of emitters, is a crucial quantum optical phenomenon with far-reaching implications in nanophotonics and many-body optical processes. Despite its observation in various systems, realizing superfluorescence in an electron-hole plasma (EHP) at room temperature has remained a formidable challenge, hindering the development of continuous-wave and electrically excited superfluorescence devices. Herein, we address this challenge by condensing the high-density EHP into an electron-hole liquid (EHL) at room temperature, thereby preserving quantum coherence. Using a model system of nanocrystals thin films, we demonstrate the first experimental observation of room-temperature superfluorescence from an EHL. Key attributes heralding superfluorescence include a redshift of ~ 94 meV from uncorrelated exciton emission, a fluence-dependent delayed growth of macroscopic coherence with abrupt radiative decay ~ 1250 times faster than spontaneous emission, a distinct quadratic fluence dependence with a clear threshold, and Burnham-Chiao ringing. These findings open up exciting possibilities for developing electrically pumped colloidal nanocrystals lasers and quantum technologies operating at room temperature.

Cooperative interactions and correlations among quantum particles represent a fascinating frontier in quantum optics and quantum technologies [1–4]. These many-body correlations underpin exotic quantum phases that surpass traditional scientific boundaries and hold the potential to unlock technological advancements across diverse fields. Notable examples include metal-insulator transitions [5], topological states [6], correlated metals [7], Bose-Einstein condensates [8], superconductivity and superfluidity [9], electron-hole liquids (EHL) [10, 11], and superfluorescence [12]. All these examples showcase the rich and complex nature of cooperative interactions at the macroscopic level, with far-reaching implications for fundamental understanding and applications.

Among these phenomena, superfluorescence holds particular significance as it combines the quantum effects of light and matter, offering promise for future hybrid optoelectronic technologies [13]. It arises from the cooperative coupling of emitters, where a large number N of quantum emitters must be confined in a volume smaller than the λ^3 (λ is the emission wavelength) [14]. In such conditions, initially uncorrelated quantum emitters spontaneously synchronize into a macroscopically correlated state. This phase-locked macroscopic coherence of N emitters produces a delayed coherent superfluorescent pulse characterized by several signatures that uniquely distinguish it from other emission processes, such as peak intensity, coherence build-up time, and decay time scaling as N^2 , $\frac{\ln N}{N}$, and N^{-1} respectively [15, 16]. Superfluorescent pulses have been observed in various platforms,

including atomic systems [17, 18], Bose-Einstein condensates [19], thin films of π -conjugated polymers [20], and selected semiconductor nanostructures [21–23] as well as crystals [19]. However, these observations were primarily made at low temperatures [15, 21, 23, 24] or high magnetic fields [21, 24]. Moreover, macroscopic quantum coherence emission at room temperature was recently observed in certain selected systems, where the macroscopic coherence is protected by using energy transfer [4], complex microcavities [25], large polarons [26] or excitons [27].

Even though superfluorescence has been observed in a wide range of systems, the development of this phenomenon based devices has not been achieved till now. This is mainly due to the excess carrier density generated in the electrical or continuous-wave pumping [28, 29] that tends to disassociate the excitons or polarons into free carriers. The resulting EHP does not support the superfluorescence due to fast dephasing, preventing coherence build-up. However, achieving superfluorescence from EHP at room temperature would provide a crucial boost to device design. While substantial progress has been made by using magnetic fields [21, 24], complex microcavities [25] and near-band-edge excitation [30], all these impose new constraints on device design. To address this challenge, we propose a novel mechanism for condensing an EHP into a quasi-thermal phase of EHL at room temperature, which can protect coherence and overcome these limitations. While EHL have been primarily reported in indirect band-gap semiconductors [31, 32] and a few direct band-gap semiconductors [33, 34] at cryogenic temperatures, restricting their use in coherent light sources. Recently, the formation of EHL at room temperature was observed in atomically thin transition metal

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dichalcogenides [11, 35]. Although enhanced fluorescence was reported from the EHL state [35], the regime of superfluorescence has not yet been achieved. The metal halide perovskites (MHP) have emerged as strong candidates due to their favorable electron-phonon interactions, which support the formation of EHL at room temperature.

In this Letter, we report the first experimental observation of superfluorescence from an EHL in a solid material at room temperature, utilizing a CsPbBr₃ thin film without any external magnetic field or optical cavity. In our experiment, we create an inverted EHL state by intense femtosecond excitation. Strong electron-phonon interactions drive the EHL formation, and the resulting macroscopic coherence radiatively emits in the form of a short pulse with a peak intensity scaling as N^2 and a characteristic duration that is three orders of magnitude shorter than uncorrelated spontaneous emission. For parameters matching our experiment, we capture the pulse build-up and decay times, as well as the qualitative presence of ringing, with Maxwell-Bloch simulations.

Using a facile hot injection method [36, 37], we have synthesized CsPbBr₃ nanocrystals with high-size uniformity. These nanocrystals were subsequently used to prepare a thin film of ~ 500 nm thickness on a micro-

scopic glass slide. Detailed information on the synthesis techniques, precise thickness measurements, optical and transmission electron microscopy images, and optical absorption spectrum is provided in the Supplemental Material (SM) [38]. The ground state optical absorption spectrum of the CsPbBr₃ thin film, shown in Fig. S2 [38] reveals, a distinct exciton onset at ~ 2.445 eV.

In our study of cooperative emission from the CsPbBr₃ thin film, we employed femtosecond photoluminescence (PL) spectroscopy. The experimental setup involved exciting the sample with an amplified laser system that generates pulses with an energy of 3.1 eV and a duration of 120 femtoseconds. By varying the excitation fluences of the laser pulses, we selectively addressed different exciton and EHP ($< 850 \mu\text{J}/\text{cm}^2$) and EHL ($\geq 850 \mu\text{J}/\text{cm}^2$) emission regimes. The latter leads us above the Mott carrier density, where excitons dissociate into free carriers and condense. Cooperative emission of the CsPbBr₃ thin film first becomes apparent in the fluence-dependence of their PL spectra at room temperature, as shown in Fig. 1(a), with cross-sectional profiles of selected fluences in Fig. 1(b).

For fluences below $50 \mu\text{J}/\text{cm}^2$, the thin film exhibited an uncorrelated spontaneous exciton emission peak centered at ~ 2.389 eV, with a full-width at half maximum (FWHM) of ~ 85 meV. As the fluence increased beyond $200 \mu\text{J}/\text{cm}^2$, a second sharp and intense emission peak emerged at ~ 2.315 eV. This peak was redshifted by ~ 74 meV from the energy of spontaneous emission and had a narrower bandwidth of ~ 30 meV, undergoing exponential decay (see Fig. S4b) [38], indicative of uncorrelated amplified spontaneous emission (ASE). In this collective phenomenon, spontaneous emission from a group of emitters is amplified, resulting in significant gain and a narrower emission bandwidth.

As the pump fluence exceeded a critical threshold ($850 \mu\text{J}/\text{cm}^2$), the PL spectrum manifested a notably sharp and intense emission peak at 2.295 eV, as shown in Fig. 1(a) and 1(b). This emission peak was redshifted by ~ 94 meV from the spontaneous emission and featured a narrower bandwidth of ~ 30 meV. Remarkably, as the fluence continued to increase, the PL intensity exhibited a scaling behavior with fluence, demonstrating a nearly quadratic relationship with an exponent of 1.8 ± 0.1 , as depicted in Fig. 1(c), while maintaining both the energy and spectral width invariant. This behavior, in which the emission intensity scales with the fluence while keeping the energy and spectral width of the PL independent of fluence, aligns with the characteristics of an EHL, echoing findings from our previous studies [39] and in other semiconductor materials [35]. Further below, we will demonstrate that this feature heralds superfluorescence from an EHL.

The formation of an EHL necessitates two primary conditions: (i) a high carrier density and (ii) a sufficiently close proximity between the carriers. When these conditions are satisfied below a critical temperature (T_c) and

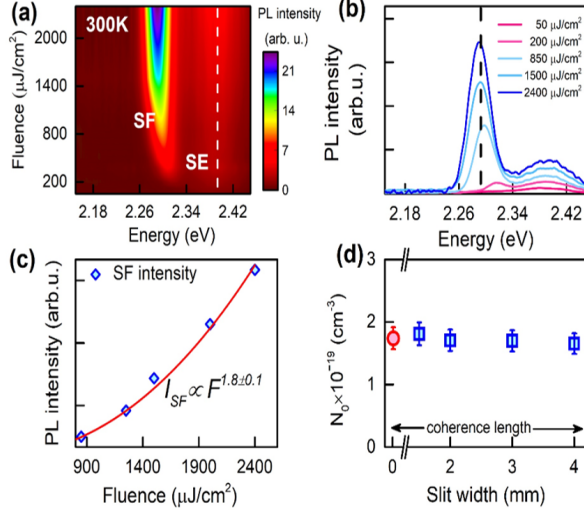


FIG. 1. (a) Fluence-dependent PL spectrum of the CsPbBr₃ thin film at room temperature, revealing the superfluorescence (SF) feature at 2.295 eV, ASE feature at 2.315 eV and an uncorrelated spontaneous emission peak (SE) at 2.389 eV. (b) PL spectra for various excitation fluences, demonstrating that at higher fluence, the position of the superfluorescence peak remains constant, reflecting the constant density of the underlying EHL. (c) Variation of PL intensity as a function of excitation fluence, showing a nearly quadratic dependence on excitation fluence. (d) The onset carrier density (N_0) for obtaining superfluorescence at various rectangular excitation lengths (blue symbols) and for circular excitation spots (pink symbol). The black horizontal arrow indicates the inferred cooperative length of the superfluorescent medium.

above an injected carrier density (n_c), the EHP undergoes a phase transition, condensing into the EHL phase. This EHL phase represents an eigenstate below the exciton energy level, preserving coherence and thus enabling room-temperature superfluorescence. An intriguing aspect of our platform is that the onset of superfluorescence in the EHL phase mainly depends on the photogenerated critical carrier density N_0 and is independent of the beam's size or shape. To demonstrate this, we employed the variable strip length method (VSLM), which also allows us to measure the cooperative length (S_c), a characteristic length upto which individual dipoles are correlated [40]. By generating a line-shaped beam of variable length utilizing a cylindrical lens, as shown in Fig. S3, we measured the onset fluence of superfluorescence as a function of strip length (S), as depicted in Fig. 1(d). Remarkably, this onset fluence remained constant for strip lengths ranging from 0.5 to 4.0 mm, mirroring the results obtained for a 0.25 mm circular spot. This behavior cannot be explained by ASE, for which the onset fluence should scale quadratically with the strip length [41]. For further details of VSLM, see Fig. S3 in [38]. From the Arecchi and Courtens cooperation time (τ_c) [40] was estimated from the VSLM as ~ 50 ps using the relation $\tau_c \geq 2nS_c/c$, where n and c represent the refractive index of the thin film [42], and speed of light. While superfluorescence decay time (τ_{SF}) ~ 0.6 - 1.4 ps from transient absorption (TA) discussed in the later section, we calculated $K\tau_c > 1$, where K is superfluorescence decay rate, a theoretical condition [12] required for superfluorescence. From the condition $K\tau_c \leq T_2^*$ (where T_2^* is dephasing time), we could roughly estimate that T_2^* is more than 50 ps at room temperature. It is important to note that the room-temperature superfluorescence observed from an EHL at high carrier densities differs significantly from the mechanism observed in hybrid perovskite films with soft polar lattices, where it originates from polarons [23, 26]. While large polarons in perovskites play a pivotal role in maintaining coherence, they become unstable at high carrier densities, especially within the context of the EHL. This instability challenges the efficacy of superfluorescence under such conditions. The significance of our findings lies not only in the elucidation of superfluorescence mechanisms in EHL but also in the broader applicability of our approach. Furthermore, our method's compatibility with other semiconductors such as transition metal dichalcogenides [35], CdS and GaAs [32], makes it highly versatile and opens pathways for exploring room-temperature superfluorescence across a wider spectrum of materials.

To provide robust experimental evidence supporting the presence of superfluorescence from EHL, we conducted an extensive investigation into carrier lifetimes and the behavior of excited correlated states. Our approach involved employing ultrafast TA spectroscopy, a well-established technique for measuring the lifetimes of superfluorescent states [21, 23, 26]. We performed TA measurements under both excitonic ($30 \mu\text{J}/\text{cm}^2$) and

EHL ($2400 \mu\text{J}/\text{cm}^2$) regimes. These regimes are associated with uncorrelated and correlated emissions, respectively. Utilizing 120 femtoseconds pump pulses centered at 3.10 eV, we monitored the change in absorbance ΔA , which represents the difference between the excited state and ground state probe absorption.

To gain a comprehensive understanding of the superfluorescence arising from the EHL phase at room temperature, we have thoroughly analyzed the fluence-dependent TA. Figure 2(a) showcases the ΔA spectra acquired at a pump-probe delay time of 1.2 ps, revealing intriguing dynamics. As the fluence increases, we observe a blueshift and broadening in the ΔA spectra. These changes saturate at $850 \mu\text{J}/\text{cm}^2$, beyond which both the bleach position and width remain nearly constant even with a further increase in influence. This striking observation can be attributed to the increased carrier density at high excitation fluences, shortening the mean distance between excitons and the corresponding enhanced Coulomb screening which reduces the exciton binding energy. Eventually, this leads to the ionization of excitons into an EHP, effectively suppressing the exciton resonance, as illustrated in Fig. 2(b) and Fig. S9 and S10 in [38]. This response strongly suggests the formation of a new many-body correlated ground state, marking the onset of the transition of the system into the EHL regime, which is also supported by the constant density behavior of PL [35, 39, 43]. These findings provide compelling evidence for the presence of superfluorescence in the EHL and offer valuable insights into the formation and behavior of correlated states.

The TA spectra of thin films at different probe delays and for a sufficiently high excitation fluence of $2400 \mu\text{J}/\text{cm}^2$ are mapped in the contour plot shown in Fig. 2(c). The most prominent features of the TA spectrum are: (1) At a probe delay time ($\Delta t < 1\text{ps}$), we observe the presence of hot carriers and a bleach feature spanning the energy range from 2.38 to 2.60 eV. Concurrently, a broad and featureless excited state absorption emerges in the lower energy region, extending from 2.18 to 2.38 eV. (2) As time progresses beyond 1 ps ($\Delta t > 1\text{ps}$), the initially generated hot carriers thermalize to the band edge and lead to a broad bleach signal. Simultaneously, the previously observed broad excited state absorption from 2.18 to 2.38 eV completely disappears. (3) A distinctive and rapidly decaying redshifted bleach signal, indicative of superfluorescence, emerges below the exciton energy within this timeframe, as indicated by the red rectangle in Fig. 2(c).

The defining characteristics of superfluorescence originating from the EHL phase in our experiments encompass the following key aspects: (1) The coherence buildup through spontaneous synchronization driven by vacuum fluctuations, representing the self-organization stage. This stage is experimentally observable as τ_B , denoting the temporal delay between the excitation pulse and the intense burst. It exhibits a $\ln F/F$ dependence

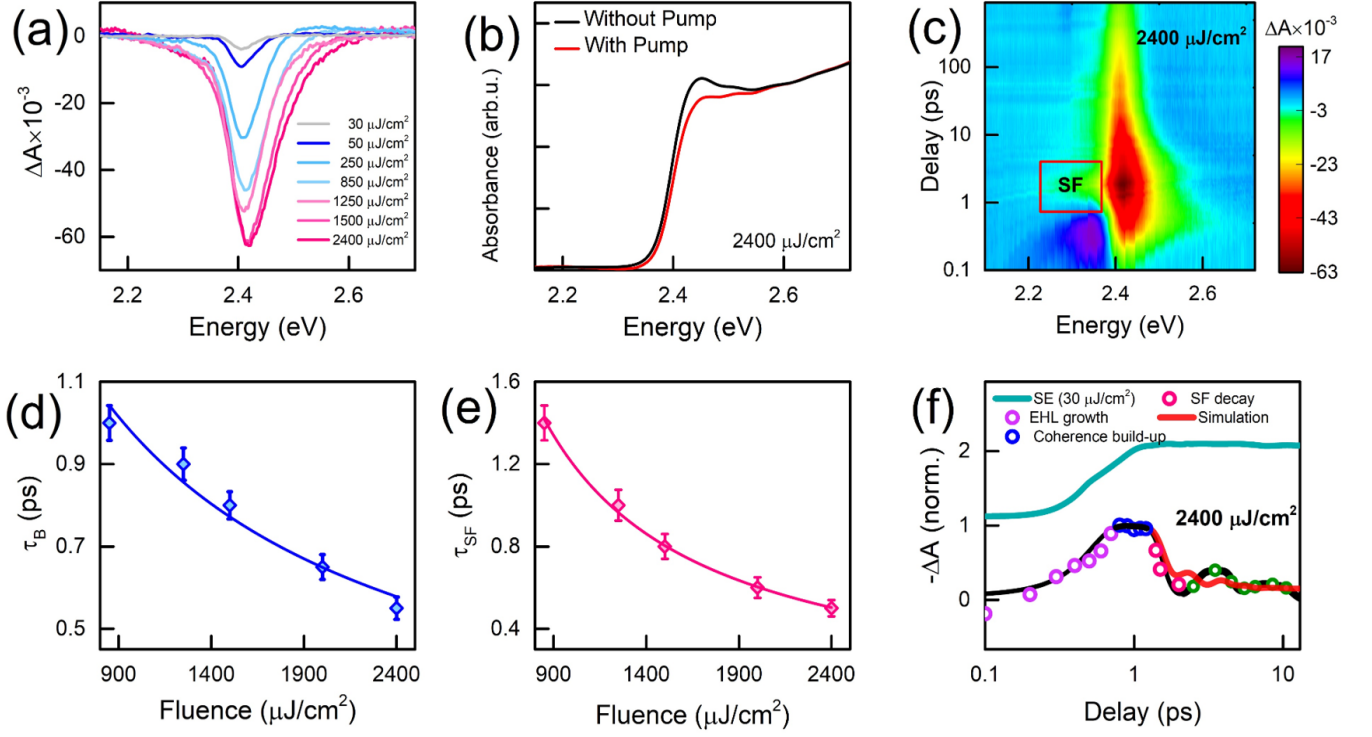


FIG. 2. (a) Spectral slices of TA at a delay time of 1.2 ps for various fluences highlighting the invariant energy of the excitonic bleach feature beyond 850 $\mu\text{J}/\text{cm}^2$. (b) Comparison of absorption spectra, without (black) and with (red) pump after 1.2 ps of photoexcitation in the superfluorescence (SF) regime at 2400 $\mu\text{J}/\text{cm}^2$, illustrating the complete suppression of the exciton resonance. (c) Contour plot of TA in the superfluorescent regime at 2400 $\mu\text{J}/\text{cm}^2$ illustrating the superfluorescence and optical gain (red square bracket). (d) Variation of the coherence build-up time (τ_B) as a function of excitation fluence (F), exhibiting the scaling characteristic for superfluorescence. (e) Superfluorescence state decay time (τ_{SF}) as a function of excitation fluence, showing a $1/F$ dependence. (f) Population decay in the superfluorescence state at 2.295 eV for 2400 $\mu\text{J}/\text{cm}^2$, purple symbols show the growth of EHL state, blue symbols show the coherence build-up, the pink symbols show the superfluorescence decay time, the green symbols show the ringing behavior, the solid black line is a fit using Eqns. (1) and the solid red line a numerical solution of the Maxwell-Bloch equations. The cyan line uncorrelated exponential decay of the spontaneous emission feature at 2.389 eV for 30 $\mu\text{J}/\text{cm}^2$. We scaled the spontaneous emission for clarity.

on excitation fluence (F), as depicted in Fig. 2(d), which is a hallmark of cooperative emission. (2) A noteworthy reduction in the decay time (τ_{SF}) of the superfluorescence at 2.295 eV, following a $1/F$ dependence, as shown in Fig. 2(e). Moreover, we observed the distinctive ringing behavior (Fig. 2f), reminiscent of radiative decay, one of the key characteristics of superfluorescence. We fit the ringing $-\Delta A(t)$ with the function:

$$-\Delta A(t) = \sum_{i=1}^3 P_i^2 \text{sech}((\tau - \tau_B)/\tau_{SF})^2. \quad (1)$$

Where P_i , represents the amplitude of each peak. To assess cooperative effects, we estimate the total number N of excited emitters participating in superfluorescence using the well-established relation [44] $N = \tau_{SE}/\tau_{SF}$ where τ_{SE} represents the lifetime of spontaneous emission. We find $\tau_{SE}/\tau_{SF} \sim 1250$, indicating that approximately $N \sim 1250$ coupled excited emitters accelerate the conventional recombination of individual excited emitters by three orders of magnitude.

We also reproduce all salient features of room temperature superfluorescence in Fig. 2(f) (red line) directly using simple simulations of the one-dimensional (1D) Maxwell-Bloch equations (MBE) [45–48], which are discussed in SM [38]. These thus show that the onset time of the super fluorescent pulse, its duration, the residual excited state population and qualitative presence of Burnham-Chiao ringing [49] are consistent with the parameters of the interpretation presented. These were an emitter density of $N_a = 1.86 \times 10^{24} \text{ m}^{-3} \simeq N/\lambda^3$ and a dephasing $T_2^* \geq 20$ ps, inferred from the VSLM. The model is also consistent with observed critical fluences at $T = 4$ and 300 K, see Figs. S11 and S12 in [38]. Residual differences between simulations and experiments are likely due to the 3D character and structure of the sample.

To provide a physical model, that can explain hitherto elusive superfluorescence at room temperature, we present a coherent 4-level model consistent with the experimental results shown in Figs. 1 and 2. The levels included represent the conduction band, the valence band, exciton states and the final EHL state, as illustrated in

Fig. S13 [38]. Initially the sample is excited with 3.1 eV energy photons, causing carriers to reach higher-lying states of the conduction band from the valence band maximum (VBM) with an excess energy of ~ 0.7 eV. This initial step gives rise to the formation of uncoupled dipoles. Subsequently, carriers undergo thermalization, gradually reaching the band edge states. As the carrier density exceeds a critical threshold density, they condense to form the EHL state with energy much below the excitonic states. For the relaxation into an EHL and stabilization of it, large electron-phonon coupling is required [33, 34]. We have calculated the electron-phonon interaction strength in terms of Fröhlich coupling constant (α) from first-principles [50–52]. The Fröhlich constant can be expressed as a product of two terms with contribution from phonons through the internal field and contrast in the dielectric screening by electrons and phonons. We estimated α and analyzed its microscopic mechanisms using mode-by-mode contributions to these two terms determined from the first principles see SM [38]. Our estimates of α of electrons and holes are 2.64 and 2.74 respectively. We find that the internal field is predominantly controlled by the LO phonon at 169 cm^{-1} through its LO-TO splitting [52], while the screening is controlled essentially by the softest of the IR-active modes at 14 cm^{-1} . Thus, we conclude that large α in CsPbBr₃ is associated with two phonons, one causing a large internal field and another screening it strongly. Their descriptors are anomalous Born charges and low phonon frequency respectively. We find that for CsPbBr₃, the value of $\alpha \sim 2$, comparable to other Pb-based halide perovskites confirms that the potential of EHL formation at room temperature is possible. Macroscopic quantum coherence spontaneously emerges within this EHL state, driven by the vacuum fluctuations as also captured by our MBE simulations. Then, the macroscopic quantum coherence gives rise to a delayed superfluorescence burst, relying on collective behavior of carriers in the EHL state.

In summary, we demonstrate a mechanism for the solid-state realization of room-temperature superfluorescence from an EHL without the need for external stimuli such as magnetic fields, complex microcavities, or low temperatures. We achieved this hallmark result by creating an inverted EHL state by intense femtosecond excitation in the CsPbBr₃ thin film. The state is formed by the condensation of carriers above a critical carrier density. The EHL then provides a protected inverted state at sufficiently high density, enabling the emergence of macroscopically quantum coherent states and the emission of superfluorescent bursts, for which we have shown clear spectroscopic signatures. The main advantage of our results is the ability of the constant density EHL to generate coherent light pulses with stable linewidth. This EHL-based superfluorescence can be utilized in advanced semiconductor technology to realize integrated photonic chip devices. Furthermore, our method offers clear advantages as it can be extended to other inor-

ganic semiconductors, such as monolayer transition metal dichalcogenides, CdS, and GaAs, where EHL has already been realized. Ultimately, these results pave the way for the development of electrically driven devices capable of producing coherent superfluorescent pulses with an on-demand wavelength at room temperature.

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