Characterizing critical phenomena via the Purcell effect

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We investigate the role of phase transitions into the spontaneous-emission rate of a single quantum emitter embedded in a critical medium. Using a Landau-Ginzburg approach, we find that in the broken symmetry phase, the emission rate is reduced, or even suppressed, due to the photon mass generated by the Higgs mechanism. Remarkably, its sensitivity to the critical exponents of the phase transition allows for an optical determination of universality classes. When applied to the cases of superconductivity and superfluidity, we show that the Purcell effect also provides valuable information on spectroscopic and thermodynamic quantities, such as the size of the superconducting gap and the discontinuity in the specific heat at the transition. By unveiling that a deeper connection between the Purcell effect and phase transitions exists, we demonstrate that the former is an efficient optical probe of distinct critical phenomena and their associated observables.

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Critical phenomena and phase transitions are among the most important and interdisciplinary research areas in physics. Criticality is known to dramatically affect many structural, thermal, and electrical properties of matter [1]. The importance of the concept of criticality extrapolates the domains of physics and finds applications in mathematics, biology, chemistry, and even economics and social sciences [1]. In addition to its phenomenological relevance, the field of critical phenomena has always been the scenario of new and groundbreaking theoretical ideas over the years, such as renormalization group and topological phase transitions [2].

In optics, critical phenomena in matter also show up in a crucial way. Examples are the optical bistability [3], the many optical manifestations of structural transitions in liquid crystals [4], and, more recently, the optical analog of the spin-glass transition in random [5,6] and homogeneous [7] lasers. The development of structured, artificial material platforms to investigate light-matter interaction, such as photonic crystals and metamaterials, has opened new venues to investigate optical manifestations of phase transitions. For instance, manifestations of the percolation phase transition were experimentally shown to occur in the Fano line shape that describes light reflection upon disordered photonic crystals [8].

The high sensitivity of the spontaneous-emission (SE) rate of an excited dipole emitter to the local environment makes the Purcell effect [9] especially prone to be influenced by phase transitions in matter. Indeed, the Purcell effect and single-molecule spectroscopy are unique tools to locally probe the electromagnetic environment at the nanoscale [10], with applications in solar cells [11], molecular imaging [10,12], and single-photon sources [13]. Of particular interest is the emergence of the new, heteroepitaxially grown, nanostructured bulk materials, such as the organohalide perovskites [14]. Here, the unique combination between the excellent electrical transport properties of the perovskite matrix and the high radiative efficiency of the colloidal quantum dots holds promise for large-scale manufacturing of infrared optoelectronic devices [15], such as multijunction solar cells and blue light-emitting diodes. In addition, progress in the field of nanophotonics and metamaterials has allowed for unprecedented control of the SE rate in artificial media such as invisibility cloaks [16], graphene-based structures [17], nanoantennas [18], photonic crystals [19], and hyperbolic metamaterials [20]. In particular, the latter may undergo a topological phase transition that manifests itself in the Purcell factor [21]. By inducing long-range spatial correlations, structural phase transitions were demonstrated to have a dramatic impact on the distribution of decay rates in disordered photonic media [22]. The SE of emitters embedded in a medium undergoing a structural phase transition induced by the temperature is also characteristically affected at criticality [23]. Another example is the percolation transition, which was shown to largely enhance the decay rate of quantum emitters and crucially govern decay pathways [24]. Fluctuations of the local density of states were experimentally shown to be maximum in thin metallic films near the percolation transition [25]. Altogether these recent findings on the Purcell effect at phase transitions, of different physical origins, suggest that a more general and profound connection between these phenomena should exist.

To elucidate this issue, in the present paper, we investigate the effects of phase transitions into the SE rate of emitters embedded in a bulk critical medium. By means of a Landau-Ginzburg (LG) description of the order parameter fluctuations, we find, without specifying *a priori* any particular physical system, that in the broken symmetry phase, the emission rate is reduced or even suppressed due to the photon mass generated by the Higgs mechanism. We show that SE presents a remarkable dependence upon the critical exponents associated to a given phase transition, allowing for an optical determination of universality classes. As concrete, realistic examples, we apply our results to the cases of a quantum emitter embedded in (i) a superconductor, either BCS or non-BCS, and (ii) superfluid ^{3}He. In both cases, we are able to extract valuable information on spectroscopic and thermodynamic quantities, such as, for example, the size of the Cooper pair order parameter and the discontinuity in the specific heat at the transition, quantities with respect to which the SE rate is highly sensitive, for either the s- or d-wave types of superconductors, as well as for p-wave pairing in superfluid ^{3}He, within the Higgs phase. Finally, in the symmetric phase, we demonstrate that critical fluctuations of the order parameter lead to an anomalously large enhancement of the SE at the critical point. Throughout the manuscript, we use MKS units when presenting our results for the SE rates and for related physical discussions, but, for simplicity, we adopt natural units $\hbar = c = k_B = 1$ during our calculations.

The Purcell effect is characterized by a modification of the SE rate, Γ, of quantum emitters by its environment. For a two-level system in free space, the SE rate is

$$\Gamma_0 = \frac{\omega_0^3 \boldsymbol{\mu}^2}{3\pi\varepsilon_0 \hbar c^3} = 2\pi \left(\frac{\omega_0 \boldsymbol{\mu}^2}{6\varepsilon_0 \hbar}\right)\rho_0(\omega_0), \tag{1}$$

where ω_0 is the two-level transition frequency, $\boldsymbol{\mu}$ is the transition dipole moment, and we identified the local density of states (LDOS) in vacuum, $\rho_0(\omega) = \omega^2/\pi^2c^3$. Changes in the electric dipole coupling and/or boundary conditions usually modify Eq. (1), which can be readily identified by rewriting the SE rate (divided by 2π) as

$$g^2\rho_0(\omega) \equiv \frac{1}{\hbar\varepsilon_0}\sum_{\lambda=\pm}\int \frac{d^3\mathbf{k}}{(2\pi)^3}|\hat{\epsilon}_{\mathbf{k},\lambda}\cdot\boldsymbol{\mu}|^2\omega_{\mathbf{k}}^2\mathcal{A}^{(0)}(\omega_{\mathbf{k}},\omega), \tag{2}$$

where we introduced a generalized DOS, $g^2\rho_0(\omega)$, $\hat{\epsilon}_{\mathbf{k},\lambda}$ is the polarization versor, $\omega_{\mathbf{k}} = c|\mathbf{k}|$, and $\mathcal{A}^{(0)}(\omega_{\mathbf{k}},\omega)$ is the diagonal part of the free-photon spectral function $\mathcal{A}^{(0)}_{\mu\nu}(\omega_{\mathbf{k}},\omega) = \eta_{\mu\nu}\mathcal{A}^{(0)}(\omega_{\mathbf{k}},\omega)$, with

$$\mathcal{A}^{(0)}_{\mu\nu}(\omega_{\mathbf{k}},\omega) = \frac{\eta_{\mu\nu}}{2\omega_{\mathbf{k}}}\{\delta(\omega-\omega_{\mathbf{k}}) - \delta(\omega+\omega_{\mathbf{k}})\}, \tag{3}$$

obtained from the free-photon propagator (we work in the Feynman gauge)

$$G^{(0)}_{\mu\nu}(k^2) = \frac{i\eta_{\mu\nu}}{k^2}. \tag{4}$$

Here, $k_\mu = (\omega/c,\mathbf{k})$, $\eta_{\mu\nu} = \mathrm{diag}(1,-1,-1,-1)$, and we used Lorentz invariance to simplify the dependence of $G^{(0)}_{\mu\nu}$ to k^2. In fact, it is easily seen that substitution of (3) into (2) leads to (1).

Equation (2) relates the SE rate directly to a property of its environment: the Green's function of the quantized electromagnetic field in free space. For interacting fields, a natural generalization of the electromagnetic contribution for the SE rate is

$$\Gamma \equiv 2\pi g^2\rho(\omega_0), \tag{5}$$

where $g^2\rho(\omega)$ is now given by an analog of Eq. (2), but with $\mathcal{A}^{(0)}(\omega_{\mathbf{k}},\omega)$ replaced by the interacting electromagnetic spectral function $\mathcal{A}(\omega_{\mathbf{k}},\omega)$.

We now can describe our system, in which a quantum emitter is embedded in a bulk critical medium undergoing a phase transition described by the Landau-Ginzburg effective Lagrangian [26],

$$\mathcal{L} = -\tfrac{1}{4}F_{\mu\nu}F^{\mu\nu} + \tfrac{1}{2}|D_\mu\varphi|^2 - a(T)\varphi^*\varphi - b(\varphi^*\varphi)^2, \tag{6}$$

where φ is a complex-scalar order parameter that couples to the electromagnetic field through the covariant derivative $D_\mu = \partial_\mu - ie^*A_\mu$ (with e^* being the coupling between the radiation and the order parameter, with dimensions of electric charge e), and $F_{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu$ is the field strength tensor. As usual,

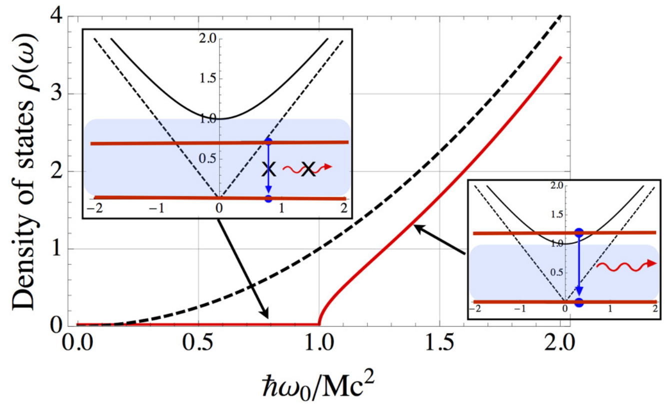

FIG. 1. Photon DOS for the vacuum (black dashed curve) and Higgs phases (solid red curve). Insets: For $\hbar\omega_0 < Mc^2$, inside the shaded area, no photons are available and no emission occurs; for $\hbar\omega_0 > Mc^2$, outside the shaded area, the DOS is finite and emission is allowed.

$a(T)$ is a function of $(T - T_c)$, where T is the temperature, and changes sign at the transition, $T = T_c$, while $b > 0$.

When $T < T_c$ and $a(T) < 0$, the φ field acquires a nonzero vacuum expectation value, $\varphi_0^2 = -a/2b = v^2$, and we need to consider perturbations around the symmetry broken vacuum, $\varphi(x) = e^{i\frac{\theta(x)}{v}}[v + \rho(x)]$, where ρ and θ describe longitudinal and transverse fluctuations of the order parameter φ, respectively. In this case,

$$\mathcal{L} = -\frac{1}{4}F_{\mu\nu}F^{\mu\nu} + \frac{M^2}{2}A_\mu^2 + \mathcal{L}_0(\rho,\theta) + \mathcal{L}_{\mathrm{int}}. \tag{7}$$

As usual, the longitudinal mode ρ becomes massive, with $m_\rho^2 = 2|a(T)|$, while the transverse fluctuations θ are massless, in accordance with Goldstone's theorem [27]. Note that a nonzero expectation value $v \neq 0$ provides the gauge field A_μ with a mass $M = ve^*$. This is the so-called Higgs mechanism [27], in which case the massive photon propagator becomes

$$G^M_{\mu\nu}(k^2) = \frac{i\eta_{\mu\nu}}{k^2 - M^2}, \tag{8}$$

so that the photon spectral function reads

$$\mathcal{A}^M_{\mu\nu}(\omega_{\mathbf{k},M};\omega) = \frac{\eta_{\mu\nu}}{2\omega_{\mathbf{k},M}}\{\delta(\omega-\omega_{\mathbf{k},M}) - \delta(\omega+\omega_{\mathbf{k},M})\}, \tag{9}$$

where the dispersion relation is $\omega_{\mathbf{k},M} = \sqrt{|\mathbf{k}|^2 + M^2}$. Calculating $g^2\rho(\omega)$ from Eq. (2) and using Eq. (5) with $\mathcal{A}^M(\omega_{\mathbf{k}},\omega)$ extracted from Eq. (9), we obtain

$$\Gamma = 2\pi\left(\frac{\omega_0\boldsymbol{\mu}^2}{6\varepsilon_0\hbar}\right)\rho_0(\omega_0)\sqrt{1 - \frac{M^2c^4}{\hbar^2\omega_0^2}} = \Gamma_0\sqrt{1 - \left(\frac{Mc^2}{\hbar\omega_0}\right)^2}, \tag{10}$$

for $\hbar\omega_0 \geqslant Mc^2$, while $\Gamma = 0$ for $\hbar\omega_0 < Mc^2$. Note that since the LDOS vanishes continuously with the gap opening (as seen in Fig. 1), the SE remains exponential with a rate given by Eq. (10). This is different from the case of band-edge modes in photonic crystals, where the exponential form for the SE rate no longer applies [28].

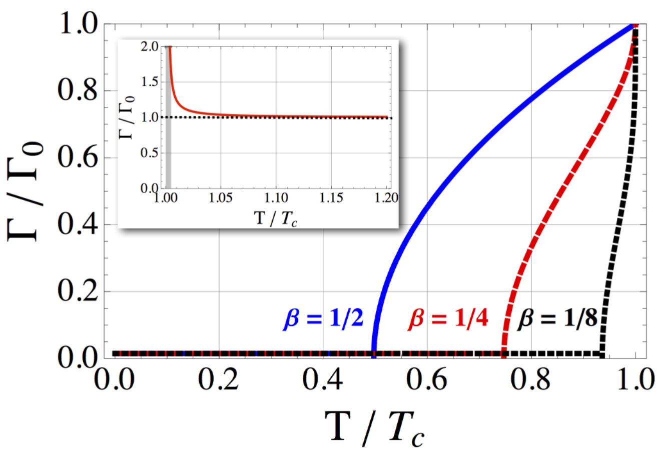

FIG. 2. Purcell factor inside the Higgs phase for different values of the critical exponent $\beta = 1/2$, $1/4$, and $1/8$, for $(M_0c^2/\hbar\omega_0)^2 = 2$. Inset: Γ/Γ_0 at the symmetric phase ($T > T_c$) for an exponential correlation length, $\xi^{-1}(T)$, with $\delta = 10$.

A nonzero photon mass reduces the value of the SE rate in the Higgs phase, and even suppresses it, for $\hbar\omega_0 < Mc^2$, when the energy $\hbar\omega_0$ is not large enough to overcome the rest energy Mc^2; see Fig. 1. This result is valid regardless of the specific form of the parameter $a(T)$, as long as it changes sign at T_c. Thus, for simplicity, but without loss of generality, we shall use a power law $M(t) = M_0|t|^\beta$ (with $t = 1 - T/T_c$), which is, in principle, valid only for $T \to T_c$. We find that in the broken Higgs phase ($T < T_c$), the SE rate increases with T and its behavior crucially depends on the value of β, as can be seen in Fig. 2 where the SE rate is shown for $\beta = 1/2$, $\beta = 1/4$, and $\beta = 1/8$. Away from criticality, the evolution of $M(t)$ will be system dependent, but nevertheless its effects on the SE rate will be present not only close to the transition ($T \sim T_c$), but for the entire $0 < T < T_c$ range, as we show below in two concrete examples: superconductivity and superfluidity.

We now focus on the behavior of the SE rate in the symmetric phase. For $T > T_c$, $a(T) > 0$ and there is no spontaneous breakdown of the vacuum symmetry, $v = 0$. This phase is described by the Lagrangian in Eq. (6), with $(m^2/2) = a(T) > 0$ for all components of φ, and the photon is massless. Thus, the presence of a medium surrounding the emitter does not lead to a position shift of the pole in the photon propagator, but it rather renormalizes the vacuum polarization [27] $i\Pi_{\mu\nu}(q^2) = iq^2\eta_{\mu\nu}\Pi(q^2)$, in such a way that

$$G^Z_{\mu\nu}(q^2) = Z(q^2)G^{(0)}_{\mu\nu}(q^2), \tag{11}$$

with $Z^{-1}(q^2) = 1 - \Pi(q^2) = 1 - [\Pi_2(q^2) - \Pi_2(0)]$.

The function $\Pi_2(q^2)$ is regular at $q = 0$, ensuring that the photon remains massless to all orders in perturbation theory, but diverges in the ultraviolet limit, $q^2 \to \infty$. For either spinorial or scalar quantum electrodynamics, one usually chooses the *on-shell* renormalization condition $\Pi(q^2 = 0) = 0$ in such a way that the residue of the photon propagator, $Z(q^2 = 0) = 1$, for the massless photon [27]. In solid-state systems, however, there are no ultraviolet divergences but, instead, there is always some scale Λ that corresponds to a given natural cutoff in the problem (e.g., the Fermi momentum k_F or the inverse lattice spacing $1/a$). Letting $q^2 = \Lambda^2 \gg m^2$, we calculate $\Pi_2(\Lambda^2) - \Pi_2(0) = \frac{\alpha^*}{12\pi}\ln(\Lambda^2/m^2)$, where $\alpha^* = (e^*)^2/4\pi$ is the fine-structure constant and $m(T) \sim \xi^{-1}(T)$ is the inverse correlation length, defined by $\langle\varphi^*(\mathbf{r}')\varphi(\mathbf{r})\rangle \sim e^{-|\mathbf{r}'-\mathbf{r}|/\xi}$. We now set $\Lambda = 1/\xi_0$, where ξ_0 is the asymptotic value of the correlation length far away from the critical point, $\xi(T \gg T_c) \approx \xi_0$, in such a way that for $T \gg T_c$, we also end up with $\Pi(q^2 = \xi_0^{-2}) = 0$ and the residue of the photon propagator is again $Z(q^2 = \xi_0^{-2}) = 1$.

The photon spectral function corresponding to the renormalized photon propagator in Eq. (11) becomes

$$\mathcal{A}^Z_{\mu\nu}(\omega_\mathbf{k};\omega) = Z(\xi^{-2})\mathcal{A}^{(0)}_{\mu\nu}(\omega_\mathbf{k};\omega), \tag{12}$$

and, as a consequence, the SE rate is given by

$$\Gamma = Z(\xi^{-2})\,2\pi\left(\frac{\omega_0\boldsymbol{\mu}^2}{6\varepsilon_0\hbar}\right)\rho(\omega_0) = Z(\xi^{-2})\,\Gamma_0. \tag{13}$$

For $T \gg T_c$, we obtain $Z \to 1$ and thus $\Gamma \to \Gamma_0$. As the critical point is approached, $T \to T_c$, the correlation length diverges, $\xi \gg \xi_0$, and $Z \gg 1$, leading to a large enhancement of the SE rate (up to the Landau pole when perturbation theory breaks down in the tiny gray area in the inset of Fig. 2).

The inset of Fig. 2 shows the normalized SE rate with an exponential form for the correlation length, $\xi(t) = \xi_0\exp(\delta/|t|)$, for $T > T_c$.

We now apply our results to the cases of a quantum emitter embedded in a superconductor, BCS, or high-T_c cuprate, and in superfluid ^{3}He. In both cases, fermions pair up to form bosons, whose fluctuations are described by an effective LG Lagrangian for a complex order parameter [29]. For superconductors, Cooper pairs of Fermi-surface electrons form singlets ($\ell = 0$ or $\ell = 2$, and $s = 0$), and the $U(1)_\varphi$-invariant effective Lagrangian is written in terms of a charged, complex-scalar field φ, which is coupled to the emitted photon A_μ, with $e^* = 2e$ [30,31]. For ^{3}He, in turn, Cooper pairs of ^{3}He atoms form triplets ($\ell = 1$ and $s = 1$), and the $SO(3)_\ell \times SO(3)_s \times U(1)_\varphi$-invariant effective Lagrangian is written in terms of not only a neutral, complex-scalar field φ, which nevertheless couples to the emitted photon A_μ, with $e^* = p_0(2\pi/\lambda)$ (with p_0 being the electric dipole moment of the ^{3}He Cooper pair and λ the wavelength of the radiation), but also in terms of $(2\ell+1)\times(2s+1)$ complex matrices Φ, describing the orbital and spin parts of the pair wave function [32]. Nevertheless, in the $U(1)_\varphi$ sector of both theories, gauge symmetry is spontaneously broken in the Higgs phase, $\varphi_0^2 \neq 0$, providing the photon A_μ with a mass M, and modifying the SE as in Eq. (10).

The use of Eq. (10) requires knowledge of both $\hbar\omega_0$ and Mc^2. Typical values for the energy splitting in semiconducting quantum dot (QDs) are of the order of $\hbar\omega_0/k_B \simeq 100$ K. The photon mass M, in turn, is proportional to the expectation value for the order parameter in the broken symmetry phase v, which is itself proportional to [33]

$$\Delta(T) = \Delta(0)\tanh\left[\pi\frac{k_BT_c}{\Delta(0)}\sqrt{\frac{2}{3}\frac{\Delta C_V}{C_N}\left(\frac{T_c}{T} - 1\right)}\right]. \tag{14}$$

Here, $\Delta(0)/k_BT_c$ is the so-called gap-to-T_c ratio and $\Delta C_V/C_N$ is the specific-heat jump at the transition, which depend on the strength of the Cooper pairing mechanism, weak or strong,

TABLE I. Values for the *gap-to-T_c* ratio, $\Delta(0)/k_B T_c$, and the specific-heat jump, $\Delta C_V/C_N$, for the considered sizes and symmetry of the order parameter ($\mathrm{OP}_{\mathrm{size}}$), from Refs. [30,32]. For the A and B phases of superfluid ^{3}He, both gaps are small and thus only the symmetry is important.

System	Superconductor		Superfluid	
$\mathrm{OP}_{\mathrm{size/sym}}$	Small gap s wave	Large gap d wave	Small gap p wave B	Small gap p wave A
$\frac{\Delta(0)}{k_B T_c}$	1.76	4.3	1.76	2.02
$\frac{\Delta C_V}{C_N}$	1.43	0.95	1.43	1.19

as well as on the topology of the order parameter, nodal or not, and for this reason we shall consider four possibilities: (i) small-gap s wave, for weakly coupled BCS, (ii) nodal, large-gap d wave, for strongly coupled high-T_c cuprates [30], (iii) small-gap p wave, for the B phase of superfluid ^{3}He, and (iv) nodal, small-gap p wave, for the A phase of superfluid ^{3}He (this one is stable at $P = 30$ bar) [32]. Typical values for these ratios, corresponding to the coupling regimes and topologies considered here, are given in Table I. For BCS superconductors, we use the upper limit of $T_c = 30$ K, valid in the weak-coupling regime, while for strongly coupled cuprates, we choose $T_c = 80$ K, as in overdoped BSCCO-2212 [30]. For superfluid ^{3}He, whose Cooper pairing occurs due to the even weaker van der Waals interaction, we shall use $T_c^A = 2.55$ and $T_c^B = 2.2$ mK [32], and $\hbar\omega_0/k_B \simeq 1$ mK, which is the typical splitting of higher (above the gap) levels in QDs. From these values of T_c, we calculate $\Delta(0)$ and, from the jump of the specific heat given in Table I, we plot Figs. 3 and 4.

As it is evident from Figs. 3 and 4, for small-gap systems such as BCS and ^{3}He, SE occurs at any temperature below T_c with a rather large Purcell factor. Conversely, for the large-gap d-wave high-T_c superconductors, the SE is suppressed for a

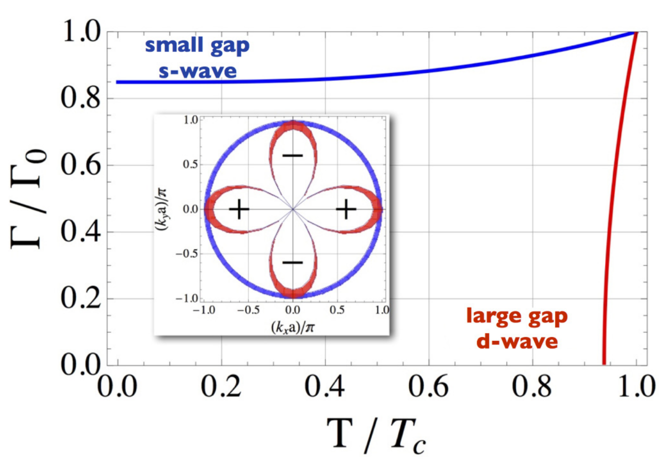

FIG. 3. Purcell factor for two limits for the size of the superconducting order parameter: (i) isotropic, small-gap s wave (in blue), and (ii) nodal, large-gap d wave (in red). Inset: Gaps in reciprocal space, normalized to their maximum value.

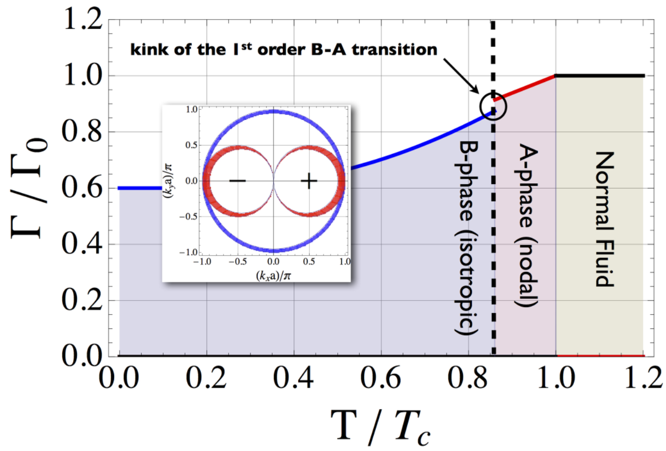

FIG. 4. Phase diagram of ^{3}He unveiled by the Purcell effect. The kink occurs at the $A - B$ transition, which separates phases with distinct Fermi-surface topologies. The behavior of Γ/Γ_0 in the symmetric phase, as $T \to T_c$, is similar to the one in Fig. 2 (not shown here due to scale limitations).

wide range of temperature and recovers only close to criticality. Remarkably, for the case of the superfluid transitions in ^{3}He, the Purcell factor captures the well-known *kink* at the transition between the two topologically distinct phases A and B [32]. Finally, for a different system other than the ones studied here, we see that by measuring the Purcell factor as in Figs. 3 and 4, we can extract the size of the gap, $\Delta(0)$, from $\Gamma(T \to 0)/\Gamma_0$ and also the jump in the specific heat, $\Delta C_V/C_N$, from the slope of $\Gamma(T \to T_c)/\Gamma_0$. This shows how an optical phenomenon, such as the Purcell effect, can provide relevant information about spectroscopic and thermodynamic quantities in critical systems.

In conclusion, we have investigated, using a Landau-Ginzburg approach, the effects of phase transitions on the SE rate of quantum emitters embedded in a critical medium. In the broken symmetry phase, we prove that the SE rate is reduced or even suppressed due to the photon mass generated by the Higgs mechanism. Remarkably, we show that one can determine critical exponents, as well as other thermodynamic and spectroscopic quantities, by means of the Purcell effect, which is revealed to be sensitive to the strength of the interaction and topology of the order parameter. This result is demonstrated in two concrete examples, namely, superconductivity (BCS or high T_c) and superfluidity in ^{3}He. Altogether, our findings suggest that the spontaneous-emission rate could be exploited as an alternative optical probe of phase transitions and their universality classes.

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