

# Long radiative lifetimes of biexcitons in GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As quantum wells

D. S. Citrin\*

*Center for Ultrafast Optical Science, The University of Michigan,  
2200 Bonisteel Boulevard, Ann Arbor, Michigan 48109-2099*

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The dynamics of biexciton radiative decay in semiconductor quantum wells is treated within the excitonic-molecule model including super-radiance, exciton recoil, and nonzero photon momentum. For parameters describing GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As quantum wells, we find that the rate for the decay of the excitonic molecule (in the ground state of the relative motion between excitons) into an exciton and a photon is less than the decay rate characteristic of radiative free one-excitons.

Nonlinear spectroscopies suggest that biexcitons play an important role in the optical response near the band gap of bulk semiconductors even at low excitation density.<sup>1</sup> It is well known that in bulk crystals the spatial correlation of the biexciton internal motion leads to the so-called giant oscillator strength, which in turn gives rise to luminescence decay times rapid compared with those characteristic of a well localized exciton.<sup>2</sup> Recent experiments on GaAs/AlGaAs quantum wells (QW's) utilizing differential transmission (DT) and time-resolved photoluminescence (TRPL) spectroscopy with resonant excitation and polarization resolution have further pointed to the importance of biexcitons.<sup>3</sup> In DT an induced absorption on the low-energy side of the heavy-hole one-exciton resonance is observed.<sup>3</sup> The induced feature displays an intensity dependence consistent with its attribution to a biexcitonic origin. The TRPL experiments reveal an extremely rapid initial decay when the pump laser and detector are cross circularly polarized.<sup>3</sup> This initial decay transient is absent in parallel polarization. The implied selection rules together with the intensity dependence indicate that the transient in the cross polarized TRPL is associated with biexcitons. In addition to the interest engendered in this fundamental topic, it is timely to have a theory of biexcitonic radiative dynamics in semiconductor QW's in order to ascertain whether the transient is indeed attributable to the intrinsic biexcitonic radiative process or to competing processes such as dissociation. We present theoretical results that indicate that the former possibility is unlikely. To carry out our analysis, we employ the widely used excitonic-molecule model (EMM) and focus on the case where the characteristic interexciton separation  $\Lambda_M$  within the biexciton is small compared with the wavelength  $\lambda$  of the emitted light in the dielectric medium — the situation for the biexciton ground state (GS) in GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW's. We show that the correlated relative motion between the constituent excitons leads to transition rates for biexciton decay into an exciton and photon which are less than the characteristic free one-exciton radiative decay rate.

The EMM assumes that a biexciton can be treated as two Wannier excitons executing correlated motion; the

internal motion within the individual excitons is taken to be the same as in the one-excitons. This model is applicable if the length scale  $\Lambda_M$  of the relative motion between excitons is large compared with the exciton Bohr radius  $a_B$ . A consequence of the EMM is that the only transition dipole present in the biexciton involves the decay of one or the other constituent excitons into a photon leaving behind a single exciton. The EMM was developed by Hanamura to describe the electronic states and radiative dynamics of biexcitons in bulk cuprous halides and II-VI materials.<sup>4</sup> Somewhat earlier, Golovin and Rashba<sup>2</sup> introduced the so-called biexciton giant oscillator strength,<sup>2</sup> based upon the earlier coherence-volume concept for localized excitons.<sup>5</sup> They argued that the radiative decay rate of the biexciton in bulk is proportional to  $\Lambda_M^3$ . The giant oscillator strength has been invoked to account for the observed rapid radiative conversion of a biexciton to an exciton in bulk crystals.<sup>6</sup>

In order to gain insight into the radiative decay of biexcitons, it is instructive to consider the decay of excitons with strongly localized center-of-mass motion due to static disorder. In this case, the concept of the coherence volume  $\Lambda^D$  ( $D$  dimensions) applied to the radiative dynamics is only valid if the scale  $\Lambda$  of the coherence is small compared with the optical wavelength in the medium of the emitted light.<sup>7</sup> In the limit of delocalized excitations,  $\Lambda \gg \lambda$ , the decay rates must approach the values given by polariton theory. For bulk, all free-exciton decay rates vanish,<sup>8</sup> while in QW's and quantum wires one finds rapid decay rates (direct-gap) for the low-lying excitonlike states near zero wave vector,<sup>9</sup> and thus the coherence-volume concept cannot be of universal applicability. In  $D = 1$  and  $2$ , the low-lying states (direct-gap) of the disorder potential have *long* decay times compared with the zero-wave-vector delocalized states.<sup>7</sup> This is because the disorder mixes a large-wave vector nonradiative component into the wave function which for the low-lying states is primarily composed of radiative small-wave-vector Fourier components. The net result is relatively slow decay of localized excitons. This explains<sup>7</sup> the leveling off of the TRPL decay time for GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW's at low temperatures at values large<sup>10</sup> compared with the radiative-recombination times

predicted by the polariton theory.<sup>11</sup>

Based on experience with bulk materials,<sup>4</sup> one might expect that for the biexciton in a QW, considerable additional phase space for the decay process is accessed due to the ability of the exciton remaining after one photon is emitted to absorb the recoil momentum. This notion, however, relies on a mistaken comparison of what is measured in TRPL on bulk and on high-quality low-dimensional systems at low temperature. In bulk, unassisted one-exciton radiative decay is forbidden;<sup>8</sup> decay is associated with defects (surfaces, impurities) or excitations (phonons, biexcitons) which break the full translational invariance as seen by the transition dipole after integrating out other degrees of freedom. The principle decay routes at low temperature are related to defects and biexcitons. Thus, the coherence-volume picture provides a natural point from which to compare biexciton and one-exciton decay rates in bulk. In low-dimensional crystals, no-phonon decay of *free* excitons is allowed and very rapid. Thus, the coherence-volume picture is misleading. In particular, we find that the transition rate for the decay of the biexciton (in the relative-motion GS) into a photon and exciton in a QW is less than the decay rate characteristic of radiative free one-excitons.

We begin by outlining the theory describing biexcitonic radiative decay accounting for the finite photon wave vector, the exciton recoil, and the quasi-two-dimensional superradiance.<sup>7,9,11</sup> Within the EMM, the biexciton wave function  $\Psi_{\mathbf{K}_{\parallel}}$  for the state with in-plane center-of-mass wave vector  $\mathbf{K}_{\parallel}$  can be represented as a superposition of free-exciton pair states. In the present study we shall concentrate on optical, rather than solid-state, phenomena. Therefore, for now we neglect the internal (including spin) degrees of freedom of the exciton. If  $\psi_{\mathbf{k}_{\parallel}}$  is the wave function for a free Wannier one-exciton with in-plane wave vector  $\mathbf{k}_{\parallel}$ , then the wave function of a free biexciton with excitation wave vector  $\mathbf{K}_{\parallel} = 2\mathbf{k}_{\parallel}$  can be expressed as  $\Psi_{\mathbf{K}_{\parallel}} = \sum_{\mathbf{k}'_{\parallel}} c_{\mathbf{k}'_{\parallel}} \psi_{\mathbf{k}_{\parallel} + \mathbf{k}'_{\parallel}} \psi_{\mathbf{k}_{\parallel} - \mathbf{k}'_{\parallel}}$  where  $c_{\mathbf{k}'_{\parallel}}$  are coefficients obtained from a calculation of the biexciton relative-motion wave function in the EMM, which give the mixing together of exciton pair states due to the correlated motion within the biexciton. Consider the decay process

$$\Psi_{\mathbf{K}_{\parallel}} \rightarrow \psi_{\mathbf{k}_{\parallel} + \mathbf{k}_{\parallel}''} + (\text{photon})_{\mathbf{k}_{\parallel} - \mathbf{k}_{\parallel}'', k_z}, \quad (1)$$

where the normal component  $k_z$  of the photon wave vector is chosen to satisfy energy conservation  $E_M(\mathbf{K}_{\parallel}) = E_{\text{ex}}(\mathbf{k}_{\parallel}) + \hbar c \sqrt{(k_{\parallel} - k_{\parallel}'')^2 + k_z^2}$  with  $c$  the speed of light in the background medium,  $E_M(\mathbf{K}_{\parallel})$  the biexciton energy, and  $E_{\text{ex}}(\mathbf{k}_{\parallel})$  the exciton energy, both measured with respect to the crystal GS. Process (1) is real if  $k_z^2 \geq 0$ . Note that (1) conserves in-plane momentum and takes into consideration the nonzero momentum carried off by the photon; the recoil momentum absorbed by the remaining exciton is automatically taken into account. Using Fermi's golden rule and summing over all final states (alternatively taking the imaginary part of the relevant radiative self-energy in the exciton-pole approximation)<sup>7</sup> gives the (amplitude) rate for process (1),

$$\Gamma_M(\mathbf{K}_{\parallel}) = \sum_{\mathbf{k}'_{\parallel}} |c_{\mathbf{k}'_{\parallel}}|^2 \left[ \Gamma(\mathbf{k}_{\parallel} - \mathbf{k}'_{\parallel}) + \Gamma(\mathbf{k}_{\parallel} + \mathbf{k}'_{\parallel}) \right], \quad (2)$$

where  $\Gamma(\mathbf{k}_{\parallel})$  is the rate for one-exciton (amplitude) decay,

$$\psi_{\mathbf{k}_{\parallel}} \rightarrow (\text{photon})_{\mathbf{k}_{\parallel}, k_z}. \quad (3)$$

The rates  $\Gamma(\mathbf{k}_{\parallel})$  are identical to those obtained in Ref. [7] from the one-exciton radiative self-energy derived in the theory of QW polaritons. Energy-momentum conservation dictates that only those one-excitons lying within the light cone,  $k_{\parallel} < \kappa = 2\pi/\lambda$ , can undergo radiative decay, i.e.,  $\Gamma(\mathbf{k}_{\parallel})$  vanishes for  $k_{\parallel} > \kappa$ .<sup>9</sup>

The remainder of this paper is concerned with an analysis of Eq. (2) in physically interesting limits. Henceforth we shall assume the squared coefficients  $|c_{\mathbf{k}_{\parallel}}|^2$  are isotropic in the QW plane, i.e.,  $|c_{\mathbf{k}_{\parallel}}|^2 = |c_{k_{\parallel}}|^2$ , as is appropriate, for example, for the relative-motion GS  $j = 0$  in a QW with in-plane isotropy and parabolic bands. This should also be a good approximation for realistic QW's since band mixing, warping, and so forth do not introduce drastically different scales into the relevant parameters. We then find that Eq. (2) can be written

$$\Gamma_M(\mathbf{K}_{\parallel}) = 2 \sum_{\mathbf{k}'_{\parallel}} |c_{\mathbf{k}'_{\parallel}}|^2 \Gamma(\mathbf{k}_{\parallel} - \mathbf{k}'_{\parallel}). \quad (4)$$

It is easy to show that the rate  $\Gamma_M(\mathbf{K}_{\parallel})$  reduces to the predictions given by polariton theory and the giant oscillator strength in the respective limits. If  $\Lambda_M \gg \lambda$  then the interexciton correlation within the biexciton is very weak on the optical length scale. This is the situation for high-energy scattering states of the relative motion. Equation (4) then assumes the limiting form  $\Gamma_M(\mathbf{K}_{\parallel}) \sim 2\Gamma(\mathbf{k}_{\parallel})$ . In this case the biexciton radiates approximately as a pair of free excitons with wave vector  $\mathbf{k}_{\parallel}$  and the decay rate per exciton is just  $\Gamma(\mathbf{k}_{\parallel})$ , the polariton result. In particular, if  $K_{\parallel} > 2\kappa$ , then the decay rate vanishes since neither one-exciton can decay into a photon and conserve both energy and momentum [process (3)]. For biexciton scattering states, the biexciton decay according to process (1) is virtually indistinguishable in TRPL from the one-exciton decay via process (3), and thus we can treat the radiative dynamics simply from the one-exciton viewpoint. In the opposite limit  $\Lambda_M \ll \lambda$ , the biexciton is strongly bound on the scale of  $\lambda$ . Then we have for Eq. (4)

$$\Gamma_M(\mathbf{K}_{\parallel}) = 2|c_{k_{\parallel}}|^2 \gamma, \quad (5)$$

where  $\gamma = \sum_{\mathbf{k}_{\parallel}} \Gamma(\mathbf{k}_{\parallel} - \mathbf{k}_{\parallel})$ .<sup>12</sup> We see in the previous expressions for  $\Gamma_M(\mathbf{K}_{\parallel})$  an overall factor of 2 for biexciton decay rates compared with those for excitons. This factor is related to dipolar superradiance.<sup>13</sup> (See below.) A more pedestrian point of view is since the biexciton contains two excitons, there is twice the probability per unit time for radiative decay compared with a one-exciton.

Equation (5) — the  $\Lambda_M \ll \lambda$  limit — is very similar (apart from the factor of 2) to what we find for the case of excitons localized by static disorder;<sup>7</sup> it is simply the

giant oscillator strength result obtained by Golovin and Rashba.<sup>2</sup> This is especially clear at  $\mathbf{K}_{\parallel} = \mathbf{0}$ ; since  $|c_0|^2 \propto \Lambda_M^2$ , we find that the so-called giant oscillator strength scales with the coherence area. We emphasize that the coherence-area result follows only if  $\Lambda_M \ll \lambda$ .<sup>14</sup>

We now consider the decay of a biexciton in the GS of the relative motion between the two constituent excitons and obtain a quantitative estimate of  $\Gamma_M(\mathbf{K}_{\parallel})$ . We assume a model wave function,  $c_{\mathbf{k}_{\parallel}} = \eta_M^{-1} \pi^{-1/2} \exp\{-k_{\parallel}^2/2\eta_M^2\}$ , which leads to closed-form expressions for the decay rate  $\Gamma_M(\mathbf{K}_{\parallel})$ . Here  $\eta_M^{-1}$  plays the role of the interexciton separation  $\Lambda_M$ . Performing the angular integration, Eq. (2) gives

$$\Gamma_M(\mathbf{K}_{\parallel}) = 2 \frac{1}{\eta_M^2 \pi} e^{-k_{\parallel}^2/\eta_M^2} \int_{k'_{\parallel} < \kappa} d^2 k'_{\parallel} e^{-k'^2_{\parallel}/\eta_M^2} \times \Gamma(\mathbf{k}'_{\parallel}) I_0(2k'_{\parallel} k_{\parallel}/\eta_M^2) \quad (6)$$

with  $I_0$  a modified Bessel function of the first kind. Equation (6) is essentially what we found for the decay rate of a localized exciton in a direct-gap QW.<sup>7</sup> The differences are the overall factor of 2 and the factor  $\exp\{-k_{\parallel}^2/\eta_M^2\} I_0(2k'_{\parallel} k_{\parallel}/\eta_M^2)$ ,<sup>7</sup> which shifts the weight in  $\Gamma_M$  toward the  $k_{\parallel}$  component of the wave function within the integral. Other than the factor of 2, this result is identical to what is found for the decay rate of the GS localized exciton in type-I indirect-gap QW's, in which case  $\mathbf{k}_{\parallel}$  is the wave vector at the dispersion minimum  $E_{\text{ex}}(\mathbf{k}_{\parallel})$ .<sup>15</sup>

Our primary interest is in GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW's. Based upon the relative spectral positions of the excitonic and biexcitonic features measured in DT, we estimate  $\Lambda_M \approx 400\text{--}500 \text{ \AA}$ .<sup>3</sup> Since  $\lambda \approx 2500 \text{ \AA}$ , the appropriate limit is  $\Lambda_M \ll \lambda$  ( $\eta_M \gg \kappa$ ). Using Eq. (6) we can set narrow bounds on  $\Gamma_M(\mathbf{K}_{\parallel})$ , namely  $e^{-\mu_M^2} \tilde{\Gamma}_M(\mathbf{K}_{\parallel}) \leq \Gamma_M(\mathbf{K}_{\parallel}) \leq \tilde{\Gamma}_M(\mathbf{K}_{\parallel})$ , where

$$\tilde{\Gamma}_M(\mathbf{K}_{\parallel}) = 2 \frac{1}{\eta_M^2 \pi} e^{-k_{\parallel}^2/\eta_M^2} \int_{k'_{\parallel} < \kappa} d^2 k'_{\parallel} \Gamma(\mathbf{k}'_{\parallel}) I_0(2k'_{\parallel} k_{\parallel}/\eta_M^2) \quad (7)$$

and  $\mu_M = \kappa/\eta_M$ . To proceed we require the  $\mathbf{k}_{\parallel}$  dependence of the one-exciton decay rates. These are obtained from QW polariton theory.<sup>7,11,16</sup> Heavy- and light-hole exciton polaritons in symmetric QW's can be classified according to the direction  $\epsilon$  of the dipole moment associated with the excitation; they are of three types:  $\epsilon = T$ ,  $\hat{\mathbf{n}}_T = \hat{\mathbf{k}}_{\parallel} \times \hat{\mathbf{z}}$ ;  $\epsilon = L$ ,  $\hat{\mathbf{n}}_L = \hat{\mathbf{k}}_{\parallel}$ ;  $\epsilon = Z$ ,  $\hat{\mathbf{n}}_Z = \hat{\mathbf{z}}$  ( $z$  is the direction normal to the QW plane).<sup>16</sup> We denote the one-exciton decay rate of state  $\epsilon, \mathbf{k}_{\parallel}$  by  $\Gamma^{(\epsilon)}(\mathbf{k}_{\parallel})$ . The  $\epsilon, \mathbf{k}_{\parallel}$ -dependent decay rates are then

$$\Gamma^{(T)}(\mathbf{k}_{\parallel}) = \frac{\kappa}{\sqrt{\kappa^2 - k_{\parallel}^2}} \Gamma_{0,\parallel}, \quad (8a)$$

$$\Gamma^{(L)}(\mathbf{k}_{\parallel}) = \frac{\sqrt{\kappa^2 - k_{\parallel}^2}}{\kappa} \Gamma_{0,\parallel}, \quad (8b)$$

$$\Gamma^{(Z)}(\mathbf{k}_{\parallel}) = \frac{k_{\parallel}^2}{\kappa \sqrt{\kappa^2 - k_{\parallel}^2}} \Gamma_{0,\perp}, \quad (8c)$$

where  $\Gamma_{0,\parallel}$  ( $\Gamma_{0,\perp}$ ) is the characteristic decay rate for in-plane (normal) directions of the dipole moment.<sup>11,17</sup> Equation (7) then gives

$$\tilde{\Gamma}_M^{(T)}(\mathbf{K}_{\parallel}) = 4\mu_M^2 e^{-k_{\parallel}^2/\eta_M^2} \beta^{-1} \sinh \beta \Gamma_{0,\parallel}, \quad (9a)$$

$$\tilde{\Gamma}_M^{(L)}(\mathbf{K}_{\parallel}) = 4\mu_M^2 e^{-k_{\parallel}^2/\eta_M^2} \beta^{-2} (\cosh \beta - \beta^{-1} \sinh \beta) \Gamma_{0,\parallel}, \quad (9b)$$

$$\tilde{\Gamma}_M^{(Z)}(\mathbf{K}_{\parallel}) = 4\mu_M^2 e^{-k_{\parallel}^2/\eta_M^2} \beta^{-1} [(1 + \beta^{-2}) \sinh \beta - \beta^{-1} \times \cosh \beta] \Gamma_{0,\perp}, \quad (9c)$$

with  $\beta = 2\mu_M k_{\parallel}/\eta_M$ . Once again we see that the limit  $\eta_M \gg \kappa$  gives the giant oscillator strength result for all  $k_{\parallel}$ . We wish to see how fast the fundamental biexciton radiative decay process can be with respect to one-exciton decay. The maxima of  $\tilde{\Gamma}_M^{(T,Z)}(\mathbf{K}_{\parallel})$  occur near  $k_{\parallel} = \kappa$ . They are approximately  $\tilde{\Gamma}_M^{(T)}(2\kappa) \sim 2e^{-\mu_M^2} 2\mu_M^2 \Gamma_{0,\parallel}$  and  $\tilde{\Gamma}_M^{(Z)}(2\kappa) \sim 2(2/3)e^{-\mu_M^2} 2\mu_M^2 \Gamma_{0,\perp}$ . Since  $\eta_M \gg \kappa$ , we have  $\tilde{\Gamma}_M^{(T,Z)}(\mathbf{K}_{\parallel}) \ll \Gamma_{0,(\parallel,\perp)}$ . For the  $L$  polariton, the maximum in  $\tilde{\Gamma}_M^{(L)}(\mathbf{K}_{\parallel})$  occurs near  $\mathbf{k}_{\parallel} = \mathbf{0}$  to give ( $\eta_M \gg \kappa$ )  $\tilde{\Gamma}_M^{(L)}(\mathbf{0}) \sim 2(1/3) 2\mu_M^2 \Gamma_{0,\parallel}$ . Thus,  $\tilde{\Gamma}_M^{(L)}(\mathbf{K}_{\parallel}) \ll \Gamma_{0,\parallel}$ . This shows that for typical parameters describing GS biexcitons in GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW's, the radiative decay rates are all less than the characteristic one-exciton decay rates; certainly we do not find substantial enhancements. Our findings show that the extremely rapid transients observed in cross polarized TRPL with resonant excitation by Wang *et al.* are not likely to be attributable simply to the fundamental biexcitonic radiative processes discussed here. Instead, the population dynamics associated with the excitons and biexcitons might play an important role, since the biexciton dissociation energy is small ( $\sim 1 \text{ meV}$ ) (Refs. 3 and 18) compared with the exciting pulse bandwidth and perhaps not much larger than  $k_B T$ . Alternatively, the observed fast decay transient may point to the breakdown of the EMM. An additional possibility is that the resonant excitation process creates a superposition of biexcitons possessing one-exciton components at small  $\mathbf{k}_{\parallel}$ , thus leading to rapid decay. In this case, the observed decay time is not the decay time of a single excitation in the usual sense.

As mentioned above, our results show that there is no additional superradiant decay for biexcitons not already accounted for in the treatment.<sup>19</sup> This is due to the prior inclusion of dipolar superradiance in the quasi-two-dimensional decay rates  $\Gamma(\mathbf{k}_{\parallel})$ . The argument is cumbersome if put in the language of Wannier excitons since the resulting decay rates will not be manifestly expressed in terms of a squared cooperativity number. Therefore, without loss of generality, we consider Frenkel excitons in order to cast the argument in the simplest possible form. Consider the biexciton state at  $\mathbf{K}_{\parallel} = \mathbf{0}$  and suppose that the GS relative-motion wave function covers a total  $C$  of molecular sites within a region whose linear dimensions are all much less than  $\lambda$ . Then, from Eq. (5) we have  $\Gamma_M(\mathbf{0}) = 2C\gamma$ .  $\gamma$  is the decay rate associated

with a one-exciton localized at a single site. For  $C = 2$ ,  $\Gamma_M(\mathbf{0}) = 4\gamma$ , exactly what we expect from the viewpoint of dipolar superradiance. One can generalize the argument to an  $N$ -exciton whose relative-motion wave function covers  $C$  sites all within  $\lambda$ . If all the one-exciton components are in phase, then the decay rate for the  $N$ -exciton into an  $(N-1)$ -exciton and a photon is  $NC\gamma(\mathbf{0})$ . For  $C = N$  we have the superradiant decay rate  $\propto N^2$ . For the Wannier  $N$ -exciton, the corresponding expression is more involved, but the underlying physics concerning superradiance is similar. This is consistent with previous findings for low-dimensional molecular systems.<sup>19</sup> Thus, we do not expect an excitation-density dependence to the fundamental decay process due to additional superradiant channels.

To conclude, we have treated the radiative conversion of a QW biexciton into an exciton and a photon within the EMM accounting for the finite photon momentum, the exciton recoil, as well as superradiant effects in QW's.

For the GS biexciton in GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW's, the rate for process (1) is less, for all biexciton center-of-mass wave vectors, than the characteristic rate for one-exciton decay via process (3). This result requires that the length scale of the relative motion between excitons within the biexciton be much less than the optical wavelength in the dielectric medium of the emitted light. This slow decay is in spite of the prior inclusion of superradiance effects.<sup>13,19</sup> Because of the slow decay of biexcitons, the concept of the giant oscillator strength is misleading as it suggests rapid radiative decay of the biexciton compared with the time scale for the decay of radiative free excitons. Furthermore, the validity of the coherence-area concept is limited to cases when  $\Lambda_M \ll \lambda$ .

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\*Electronic address: citrin@mich1.physics.lsa.umich.edu

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