

Does the Phenomenological Approach Contradict the Quantum Theory of Exciton–Polariton Spatial Dispersion?¹

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Abstract—This paper clarifies the controversial issue over 40 years between the quantum approach by Pekar and the phenomenological approach by Ginzburg about the exciton–polariton spatial dispersion theory. For an isotropic nongyrotropic medium, the analytical explicit function of the impermeability tensor $\eta_{ij}(\omega, \vec{k})$ (the inverse of the permittivity tensor) is obtained from the anisotropic undamped-wave harmonic oscillator model. After expanding $\eta_{ij}(\omega, \vec{k})$ with respect to small parameters within it rather than to wavevector \vec{k} , the approximate refractive indices can be determined from the eigenvalue equation. By this treatment, the phenomenological approach is proved to be the approximation of the quantum approach near the resonance frequency. The condition for the approximation is discussed.

INTRODUCTION

Exciton–polaritons are also called normal waves of electromagnetic waves in the field of optical physics. The term exciton–polariton is used more frequently in condensed-matter physics, while it is near nothing new but just another term from the optical point of view. Here, we use the terms exciton–polaritons and normal waves interchangeably.

Since the pioneer theoretical works of Pekar [1, 2] and Ginzburg [3], especially the theoretical and experimental work by Hopfield and Thomas [4] that conveyed much of the relevant physics of spatially dispersive media in a transparently clear way, the exciton–polariton spatial dispersion has been a well-developed and well-documented [5–15] subject. The investigation development of the exciton–polaritons in quantum-confined spatially dispersive systems such as GaAs quantum wells [16], as well as in other nanostructures [17], shows the potential for applications of the subject to ultrasmall optoelectronic devices.

Pekar was the first to discuss the phenomenon of optical waves in the region of a particular exciton absorption frequency in media and to shape them with the quantum theory. However, Ginzburg first connected the phenomenon with the spatial dispersion effect and reshaped it with a phenomenological approach. It has been considered [5, 6, 9, 13] for more than 40 years that Pekar’s treatment based on the microscopic quantum theory [2, 9] and Ginzburg’s treatment based on the macroscopic phenomenological approach [3, 10]

would have been different in many of their essential conclusions. Pekar concluded that [6] Ginzburg’s phenomenological treatment “leads to consequences (refractive indices) that, generally speaking, are in contradiction to the results of the quantum-mechanical investigation and also inconsistent with experiment,” and that (see [9, p. 122]) “the existence of the additional waves cannot be obtained by means of the phenomenological procedure.” Ginzburg defended himself against Pekar’s argument, but, because his defense [11] lacked academic evidence and, therefore, persuasion, even in 1998 there still was a claim that the two theories would have differed substantially [13], and the controversy has not been clarified so far. To clarify this controversy is our purpose in this paper.

PHENOMENOLOGICAL RESULT VS. QUANTUM RESULT

An essential part of Ginzburg’s phenomenological treatment is a phenomenological expansion of the impermeability tensor $\epsilon_{ij}(\omega, \vec{k})$, the inverse of the permittivity tensor $\epsilon_{ij}(\omega, \vec{k})$, in powers of wavevector \vec{k} [3] (see also Section 3.1.2 in [10]),

$$\eta_{ij}(\omega, \vec{k}) = \eta_{ij}(\omega) + i\kappa_{ijl}(\omega)k_l + \beta_{ijlm}(\omega)k_l k_m, \quad (1)$$

where all of the coefficients $\kappa_{ijl}(\omega)$ vanish for nongyrotropic crystals. The weakness in Ginzburg’s approach, however, is that, although he assumed all of the coeffi-

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icients in expansion (1) to be dependent on frequency ω , he did not know how this dependence arose; therefore, he dealt with all of the coefficients except $\eta_{ij}(\omega)$ as constants [3] (see also Chapter 4 in [10]), which is unreasonable from both the physical and mathematical points of view. Although, as we will see in this paper, $\beta_{ijlm}(\omega)$ can at last be proved to be approximately independent of frequency near the resonance frequency, the treatment of them as constants in the beginning is vulnerable to be criticized, as done by Pekar [6]. To overcome this shortcoming, we should first find a way to obtain the analytical explicit function of $\epsilon_{ij}(\omega, \vec{k})$, and a start point for this work is the anisotropic undamped-wave harmonic oscillator model to formulate a linear relation between the polarization \vec{P} and the electric field \vec{E} , suggested first by Agranovich and Kaganov [7], which reads (see also Section 4.5.1 in [10])

$$\begin{aligned} & \rho_{ij} \frac{\partial^2 P_j(t, \vec{r})}{\partial t^2} + \nu_{ij} P_j(t, \vec{r}) \\ & + \gamma_{ijl} \frac{\partial P_j(t, \vec{r})}{\partial x_l} + \mu_{ijlm} \frac{\partial^2 P_j(t, \vec{r})}{\partial x_l \partial x_m} = \Lambda_{ij}^{(0)} E_j(t, \vec{r}), \end{aligned} \quad (2)$$

where the tensors ρ_{ij} , ν_{ij} , γ_{ijl} , and others are practically independent of ω near the individual exciton dipole absorption frequency, and they are determined by the type of crystal symmetry. The last two terms on the left-hand side of Eq. (2) take into account spatial dispersion, which originates from the direct interaction between the oscillators permitting energy transmission that is not electromagnetic in origin [4, 18], and the first two terms, as well as the term on the right-hand side, have the same physical meaning with the counterparts in the isotropic harmonic oscillator model in the absence of spatial dispersion [19–21]. The media are considered ideal, so that the dissipation term $\sigma_{ij} \partial P_j / \partial t$ is not included in Eq. (2). It is clear that $\gamma_{ijl} = 0$ for nongyro-tropic media. Considering the harmonic solution of $E(t, \vec{r}) = E(\omega, \vec{k}) \exp[i(\vec{k} \cdot \vec{r} - \omega t)]/2 + \text{c.c.}$ and $P(t, \vec{r}) = P(\omega, \vec{k}) \exp[i(\vec{k} \cdot \vec{r} - \omega t)]/2 + \text{c.c.}$, where $\vec{k} = k_0 \hat{k}$, n is the refractive index, $k_0 = \omega/c$, and \hat{k} is the unit vector of \vec{k} , from Eq. (2) we find the explicit form of the permittivity tensor $\epsilon_{ij}(\omega, \vec{k})$, taking into consideration spatial dispersion

$$\epsilon_{ij}(\omega, \vec{k}) = (\epsilon_r)_{ij} + 4\pi \Delta_{il}^{-1}(\omega, \vec{k}) \Lambda_{lj}^{(0)}, \quad (3)$$

where $(\epsilon_r)_{ij}$ is the background permittivity, and

$$\Delta_{ij}(\omega, \vec{k}) = -\rho_{ij} \omega^2 + \nu_{ij} + i\gamma_{ijl} k_l - \mu_{ijlm} k_l k_m. \quad (4)$$

Now, let us take into account isotropic nongyro-tropic media, the simplest crystals with the highest symmetric property. For isotropic media, we have

$$\begin{aligned} (\epsilon_r)_{ij} &= \epsilon_r \delta_{ij}, \quad \rho_{ij} = \rho \delta_{ij}, \quad \nu_{ij} = \rho \omega_{tr}^2 \delta_{ij}, \\ \Lambda_{ij}^{(0)} &= \frac{\rho \omega_0^2}{4\pi} \delta_{ij}, \quad \gamma_{ijk} = 0, \end{aligned} \quad (5)$$

$$\mu_{ijlm} k_l k_m = \rho \mu_{tr} k^2 \delta_{ij} + \rho (\mu_{lo} - \mu_{tr}) k_i k_j,$$

where $\rho \mu_{lo}$ and $\rho \mu_{tr}$ are two independent components of μ_{ijlm} for isotropic media (see Section 4.5.2 in [10]). Introducing Eq. (5) into Eqs. (4) and (3), we obtain the analytical expression for the permittivity tensor

$$\epsilon_{ij}(\omega, \vec{k}) = \epsilon_r \delta_{ij} + (\epsilon_{lo} - \epsilon_r) \hat{k}_i \hat{k}_j, \quad (6)$$

where ϵ_{tr} and ϵ_{lo} are the transverse and longitudinal parts of $\epsilon_{ij}(\omega, \vec{k})$, respectively,

$$\begin{aligned} \epsilon_{tr} &= \epsilon_r - \frac{\omega_0^2}{\omega^2 + k^2 \mu_{tr} - \omega_{tr}^2}, \\ \epsilon_{lo} &= \epsilon_r - \frac{\omega_0^2}{\omega^2 + k^2 \mu_{lo} - \omega_{tr}^2}, \end{aligned} \quad (7)$$

and ω_{tr} is the particular resonance frequency (the transverse frequency). Furthermore, the inverse of $\epsilon_{ij}(\omega, \vec{k})$ can be got

$$\begin{aligned} \eta_{ij}(\omega, \vec{k}) &= \frac{\delta_{ij}}{\epsilon_r} \\ &+ \frac{\omega_0^2}{\epsilon_r^2 (\omega^2 - \omega_{lo}^2)} \left(\frac{\delta_{ij} - \hat{k}_i \hat{k}_j}{1 + \Omega_{tr}} + \frac{\hat{k}_i \hat{k}_j}{1 + \Omega_{lo}} \right), \end{aligned} \quad (8)$$

where $\Omega_{tr} = k^2 \mu_{tr} / (\omega^2 - \omega_{lo}^2)$, $\Omega_{lo} = k^2 \mu_{lo} / (\omega^2 - \omega_{lo}^2)$, and $\omega_{lo} = (\omega_{tr}^2 + \omega_0^2 / \epsilon_r)^{1/2}$ is the longitudinal frequency. It is obvious that an isotropic medium must, in general, be characterized by a tensorial rather than a scalar permittivity as a consequence of spatial dispersion. For the specific configuration that \vec{k} is directed along the z axis of the coordinate system, Eq. (6) degenerates as

$$\epsilon_{xx} = \epsilon_{yy} = \epsilon_{tr}, \quad \epsilon_{zz} = \epsilon_{lo}, \quad \epsilon_{ij} = 0 \quad (i \neq j). \quad (9)$$

On the other hand, Hopfield and Thomas [4] have obtained a semiphenomenological model of the scalar permittivity in the specific configuration that the polarization and electric field are parallel and both perpendicular to \vec{k} . As a matter of fact, their model is only ϵ_{xx}

or ϵ_{yy} , the transverse part of $\epsilon_{ij}(\omega, \vec{k})$. Comparing ϵ_{tr} in Eq. (7) with their model (see Eq. (9) in [4]) and the model without spatial dispersion [19–21], we have

$$\omega_0^2 = \frac{4\pi e^2 N_p f_p}{m}, \quad \mu_{tr} = -\frac{\hbar \omega_{tr}}{m_e^*}, \quad (10)$$

where N_p is the number of molecules per unit volume with Z electrons per molecule; f_p is the number of electrons per molecule with binding frequency ω_{tr} , the oscillator strength, satisfying the sum rule $\sum f_p = Z$; e is the charge of the electron; m is the rest mass of the electron; and m_e^* is the effective mass of the exciton.

Having the analytical explicit expression for $\eta_{ij}(\omega, \vec{k})$, we can further expand it with small parameters within it. First, it is reasonable to assume that μ_{tr} and μ_{lo} have the same order; then, Ω_{tr} and Ω_{lo} also have the same order. With the condition

$$\left(|\Omega_{tr}| = \left| \frac{k_0^2 \mu_{tr} n^2}{\omega^2 - \omega_{lo}^2} \right| \right) \ll 1 \quad (11)$$

(now $|\Omega_{lo}| \ll 1$ also, vice versa), Eq. (8) can be expanded with respect to Ω_{tr} and Ω_{lo} as

$$\begin{aligned} \eta_{ij}(\omega, \vec{k}) &= \eta_0 \delta_{ij} \\ &+ \beta_{tr}(k_i k_j - k^2 \delta_{ij}) - \beta_{lo} k_i k_j + o(\Omega_{tr}) \end{aligned} \quad (12)$$

where

$$\begin{aligned} \eta_0 &= \frac{\omega^2 - \omega_{tr}^2}{\epsilon_r(\omega^2 - \omega_{lo}^2)}, \quad \beta_{lo} = \frac{\mu_{lo} \omega_0^2 / \epsilon_r^2}{(\omega^2 - \omega_{lo}^2)^2}, \\ \beta_{tr} &= \frac{\mu_{tr} \omega_0^2 / \epsilon_r^2}{(\omega^2 - \omega_{lo}^2)^2}. \end{aligned}$$

Although here $\eta_{ij}(\omega, \vec{k})$ is expanded with respect to the small parameters Ω_{tr} and Ω_{lo} , rather than with respect to \vec{k} about $\vec{k} = 0$, it can be found, as a matter of fact, that the result will be the same with expansion (12) provided that Eq. (8) is expanded with respect to \vec{k} , following Ginzburg's idea [3, 10]. Then, comparing Eq. (12) with the concrete form of expansion (1) for

isotropic media (see Section 3.2.2 in [10]), we can obtain

$$\begin{aligned} \eta_{ij}(\omega) &= \eta_0(\omega) \delta_{ij}, \\ \beta_1(\omega) &= -\beta_{lo}(\omega), \quad \beta_2(\omega) = -\beta_{tr}(\omega), \end{aligned} \quad (13)$$

where $\beta_1 = \beta_{xxxx}$ and $\beta_2 = \beta_{xxxx}$ are two independent components of β_{ijkl} for isotropic media. Of course, $\kappa_{ijl} = 0$ as mentioned above. Clearly, β_1 and β_2 are functions of ω , rather than constants, but near the resonance we have $\omega^2 - \omega_{lo}^2 \approx \omega_{tr}^2 - \omega_{lo}^2 = -\omega_0^2 / \epsilon_r$; β_2 is then reduced to a constant β , i.e., is independent of frequency ω ,

$$\beta = \beta_2(\omega_{tr}) = \frac{\hbar \omega_{tr}}{\omega_0^2 m_e^*}, \quad (14)$$

which is just what was obtained by Ginzburg (Eq. (4.2.9b) in [10]).

Now, directly quoting the result for refractive indices obtained by Ginzburg [3] (see also Eq. (4.2.6) in [10]) in the coordinate system whose z axis coincides with \vec{k} , we have

$$n_{\pm}^2 = -\frac{\eta_0}{2k_0^2 \beta} \pm \sqrt{\frac{1}{4} \left(\frac{\eta_0}{k_0^2 \beta} \right)^2 + \frac{1}{k_0^2 \beta}}. \quad (15)$$

The results above can be extended to the cases of cubic nongyrotropic media belonging to the two classes $\bar{4}3m$ and $m\bar{3}m$, the cubic crystals with highest property in the cubic system, as long as the wavevector \vec{k} is directed along $[0, 0, 1]$ (or $[1, 0, 0]$, $[0, 1, 0]$) (see [10, p. 169]).

Using Eq. (10) and $\omega \approx \omega_{tr}$, we can obtain near the resonance

$$\mu_p = \frac{-\eta_0}{k_0^2 \beta} \approx C_p \left(1 - \frac{\omega_{tr}}{\omega} \right), \quad b_p = \frac{1}{k_0^2 \beta} \approx \frac{8\pi m_e^* c^2 a_p}{\hbar^2 \omega_{tr}^3}, \quad (16)$$

where

$$C_p = \frac{2m_e^* c^2}{\hbar \omega_{tr}}, \quad a_p = \frac{e^2 \hbar N_p f_p}{2m}. \quad (17)$$

However, Pekar got the following result on the refractive index in cubic crystals more than 40 years ago:

$$n_{\pm}^2 = \frac{1}{2}(\mu_q + \epsilon_r) \pm \sqrt{\frac{1}{4}(\mu_q - \epsilon_r)^2 + b_q}. \quad (18)$$

Here, μ_q and b_q have the same expressions with μ_p and b_p in Eq. (16) near the resonance frequency [2]. Using

the typical concrete data presented by Pekar [2] (see also Section 12 in [9]), we have $b_p \approx 5.8 \times 10^4$, $C_p \approx 5.1 \times 10^5$, and $\epsilon_r = 2$. Therefore, although the results on the refractive index from the phenomenological and quantum approaches appear different, when C_p and b_p are very much larger than ϵ_r the two results on the refractive index are identical near the resonance.

A CRITERION: MATERIAL CHARACTERISTIC PARAMETER α_c

It goes without saying that the error between the quantum result (18) and the phenomenological one (15) will occur both if ω is detuned away from the resonance and if C_p and b_p are not larger enough than ϵ_r even in the resonance. To discuss this issue quantitatively, we use condition (11) to find the frequency region where expansion (12) holds. Because the n_- (upper) exciton-polariton branch cannot propagate throughout the bulk crystal near the resonance [8, p. 83], only the case for n_+ (lower) branch is discussed here. In order to discuss the issue, condition (11) is rewritten as its equivalent form

$$\frac{k_0^2 \mu_{tr} n^2}{\omega^2 - \omega_{lo}^2} = \alpha, \quad (19)$$

where α is a constant. When $|\alpha| \ll 1$, Eq. (19) is just the expansion condition (11). Supposing α_{\max} ($0 < \alpha_{\max} < 1$) is the upper limit of the expansion condition (11) and introducing n_+ in Eq. (18) into Eq. (19), we have the corresponding ω_{\max} for the case that $m_e^* > 0$ ($\mu_{tr} < 0$) (only positive effect mass has been treated because it is the usual case for exciton resonances in direct band gap semiconductors),

$$\omega_{\max}^2 - \omega_{tr}^2 = \frac{k_0^2 \mu_{tr} \epsilon_r}{\alpha_{\max}} \left(1 - \frac{\alpha_{\max}^2}{\alpha_c} \right), \quad (20)$$

where $k_0 \approx \omega_{tr}/c$,

$$\alpha_c = \frac{\epsilon_r^2}{b_p} = \frac{\hbar \epsilon_r \omega_{tr}^2}{2 \omega_{LT} c^2 m_e^*}, \quad (21)$$

and $\omega_{LT} = \omega_{lo} - \omega_{tr}$, given by $\omega_{LT} \approx \omega_0^2 / (2\epsilon_r \omega_{tr})$, is the longitudinal-transverse splitting. As will be seen, α_c in Eq. (21) is a critical parameter determined uniquely by the characteristic parameters of the material. The frequency region where the expansion (12) holds for the condition $|\Omega_{tr}| < \alpha_{\max}$ is the interval $(-\infty, \omega_{\max})$. From Eq. (20), it can be derived that

$$\frac{\omega_{\max} - \omega_{tr}}{\omega_{LT}} = \alpha_{\max} \left(1 - \frac{\alpha_c}{\alpha_{\max}^2} \right) < \alpha_{\max} (< 1). \quad (22)$$

The inequality (22) is obtained because $\alpha_c > 0$ and gives another inequality $\omega_{\max} - \omega_{tr} < \omega_{lo} - \omega_{tr}$, which tells us that, no matter what materials are, ω_{\max} will always be less than ω_{lo} ; that is, expansion (12) exists only at the lower end of the longitudinal frequency ω_{lo} for the n_+ branch. ω_{lo} is a singularity of Eq. (15), and n determined by Eq. (15) is completely unreliable at the higher end of ω_{lo} . From Eq. (22), taking $\alpha_{\max} = 0.1$ (a general case), we also conclude that, when $\alpha_c > 0.01 (= \alpha_{\max}^2)$, then $\omega_{\max} < \omega_{tr}$ and ω_{\max} is located at the lower end of the resonance frequency; otherwise, $\omega_{\max} > \omega_{tr}$ and ω_{\max} is at the higher end of the resonance frequency. For the case that $\alpha_c > 0.01$ ($\omega_{\max} < \omega_{tr}$), the truncated error in expansion (12) cannot be neglected as ω gets close to ω_{tr} . The approximate n_+ from Eq. (15) will introduce neglected-less error even in the interval $(-\infty, \omega_{\max})$, because β is the value of $\beta_2(\omega)$ at $\omega = \omega_{tr}$. In contrast, when $\alpha_c < 0.01$, $\omega_{tr} \in (-\infty, \omega_{\max})$, and Eq. (15) is a good approximation of Eq. (18) in the interval. Therefore, the critical parameter α_c of a material is a criterion to judge whether Eq. (15) can approximate Eq. (18) well or not for the material. The bigger the material's α_c , the larger the error between the two results. To illuminate the conclusion, we give the comparison of the two results for three concrete materials with different α_c , as shown in Fig. 1. For the fictitious material given by Pekar [2] (see also Section 12 in [9]), its α_c is about 3×10^{-5} ; therefore, a much better approximation can be obtained.

A remark is given at the end of the paper. The microscopic quantum theory of spatial dispersion is the most rigorous but obscure [4]. The rudiments of the theory with less rigorous but without loss of physical essence are needed from the applicable and technical point of view. Such rudimentary models available now are the phenomenological one suggested by Ginzburg [3] and semiphenomenological one by Hopfield and Thomas [4]. The Hopfield-Thomas model allows one to obtain the same results with the quantum model, but their scalar permittivity can be used only in a very specific configuration (the condition of the model can be found in the paragraph above Eq. (4) in [4] and the first two paragraphs of Section 3 in the same paper). However, the expansion of $\eta_{ij}(\omega, \vec{k})$ has the advantage of making it possible to obtain an approximate analytical solution for an arbitrary configuration. As a matter of fact, the phenomenological approach was used to prove that exciton-polaritons propagate along the [1, 1, 0] and [1, 1, 1] directions in cubic crystals (see Section 4.3.1 in [10]). Therefore, in our point of view, the phenomenological description has its potential merit, especially in applicable and technical problems (when the spatial

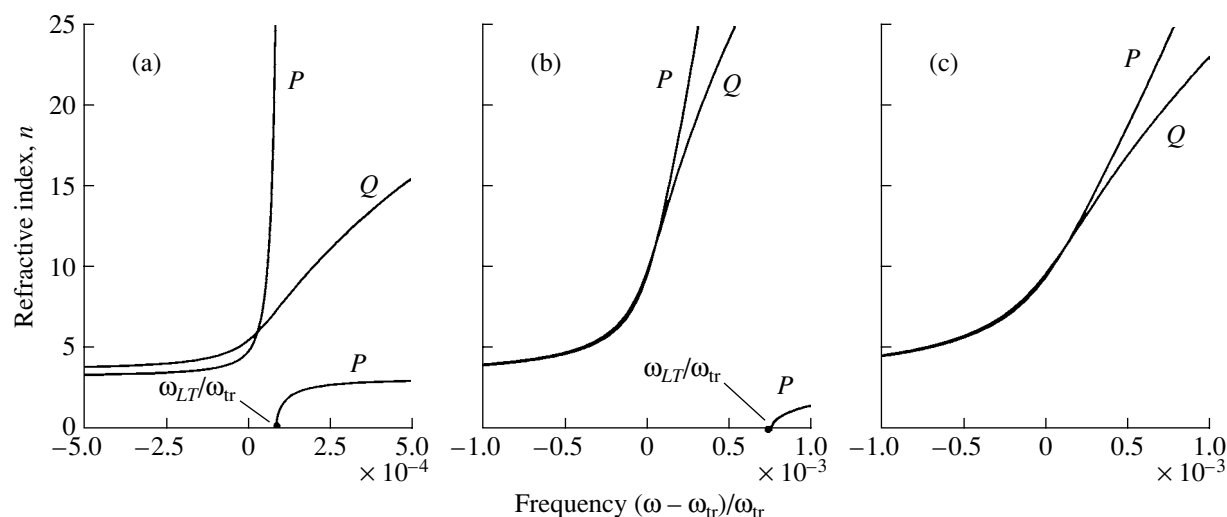


Fig. 1. Comparison of the approximate and exact refractive indices for three concrete materials with different α_c , where curve P is the result from the phenomenological approach, and curve Q is the result from the quantum one. (a) GaAs: $\alpha_c \approx 0.3$; (b) CdS: $\alpha_c \approx 0.01$; (c) HgI₂: $\alpha_c \approx 0.006$. All data come from Table 2 (p. 114) in [8]. ω_{0} (ω_{LT}) is also indicated, except in panel c (outside the frequency region drawn). ω_{0} is a singularity of the phenomenological refractive index that is completely unreliable at the higher end of ω_{0} .

dispersion theory is put into practice), although it is only an approximation of the “exact” solution along the principal axes.

CONCLUSIONS

We clarified the controversy on the exciton–polariton spatial dispersion theory between the quantum point of view and the phenomenological point of view over 40 years. The result from the phenomenological approach does not contradict that from the quantum approach but is an approximation of it. The approximate extent is determined by the material’s characteristic parameter α_c defined by Eq. (21). The greater is the material’s α_c , the larger is the error between the phenomenological result and the quantum result.

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