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Quantum entanglement based on surface phonon polaritons in condensed matter systems

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Surface phonon polariton (SPhP) is a special propagation mode in condensed matter systems. We present an investigation on the entanglement of SPhP modes. The entangled SPhP pairs are generated through launching entangled photons onto the grating coupling systems. The interaction Hamiltonian for the coupling process between entangled photons and entangled LRSPhPs is derived. State vector of the entangled LRSPhPs is obtained through the perturbation theory. The origin of LRSPhP entanglement is revealed. Wave mechanics approach is taken to describe the coupling process as an alteration. To present the nonlocality, the second-order correlation function is studied. *Copyright 2013 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License*. [http://dx.doi.org/10.1063/1.4802887]

I. INTRODUCTION

Quantum entanglement has become one of the most attractive topics during the past two decades. The quantum entanglement systems are described by entangled states that could not be decomposed into products of single particle states.[1] Usually, the entanglement property of the multi-particle system is demonstrated by entangled photon states.[2] In recent years, various schemes which investigate quantum entanglement in non-photon systems are proposed and attract plenty of attention. For instance, entanglement systems based on metallic microstructure devices which could support surface plasmon polariton (SPP) mode are investigated for effective miniaturization.[3,4] Moreover, entanglement in condensed matter systems based on phonon modes is also of great interest. Several related interesting results have been reported.[5,6]

In this paper, we present an investigation of quantum entanglement in condensed matter systems. Unlike the traditional ones[5,6] in which the entangled phonon states are generated by Raman scattering, the entangled states which we investigate in this work are surface phonon polariton (SPhP) pairs. The SPhP mode is a transverse magnetic (TM) mode vibration resulting from the coupling of an infrared photon (TM mode) with a transverse-optic (TO) phonon.[7] For classical phonon modes in condensed matter systems, thermal fluctuation usually causes serious interference in measurement and destroys the quantum coherence.[6] As an advantage, SPhP does not face this difficulty because it could be regarded as a surface electromagnetic wave in a general view.[8] The typical wavelength of SPhP is around 10 μm. The properties of SPhP are analogous to those of SPP in the near-infrared band, so it's a suitable candidate for integration circuit, especially in the mid-infrared band. The interaction Hamiltonian for the coupling process between entangled photons and entangled LRSPhPs is derived, and the state vector of the entangled LRSPhPs is obtained based on it. The entanglement

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is revealed to transmit from photon pairs to LRSPhP pairs in a low dimension spatial correlation system. Moreover, the total coupling process is further analyzed. Three types of entangled states arise from the process, i.e., entangled LRSPhP state, entangled photon state and hybrid entangled state of LRSPhP with photon. Each of them corresponds to a certain state probability. The second-order correlation of the system is also studied. Investigating these features should be helpful in revealing the fundamental mechanism of quantum entanglement.

II. ENTANGLEMENT OF SURFACE PHONON POLARITON

For a lossy, linear, isotropic polar dielectric material, the dispersion relation of SPhP mode is shown in the following formulation[9]

$$\beta^2 = \frac{\omega^2}{c^2}\left(\frac{\varepsilon_d \varepsilon_c}{\varepsilon_d + \varepsilon_c}\right), \tag{1}$$

where β is the propagation constant of the surface mode. ε_{d} and ε_{c} are dielectric constants of the two materials located at both sides of the interface, respectively. ε_{c} represents the relative permittivity of the lossy polar dielectric material that is given by the formulation[10]

$$\varepsilon_c(\omega) = \varepsilon_c(\infty)\left[1 + \frac{\omega_{LO}^2 - \omega_{TO}^2}{\omega_{TO}^2 - \omega^2 - i\gamma\omega}\right]. \tag{2}$$

ω_{LO} and ω_{TO} are the frequencies of the longitudinal optical phonon and the transverse optical phonon, respectively. And the term $\mathrm{i}\gamma\omega$ represents the damping. Usually, γ is much smaller than ω; so for frequencies falling between the TO and LO frequencies, the dielectric constant have a negative real part and a positive imaginary part. Thus the propagation constant also shows a similar form, just like that of the surface plasmon polariton. For a detailed illustration, we take SiC system as an example. Corresponding dielectric permittivity and dispersion relation of SiC are shown in Fig. 1(a) and Fig. 1(b), respectively. Related parameters are given as $\omega_{\mathrm{L}} = 969\ \mathrm{cm}^{-1}$, $\omega_{\mathrm{T}} = 793\ \mathrm{cm}^{-1}$, $\Gamma = 4.76\ \mathrm{cm}^{-1}$, $\varepsilon_\infty = 6.7$.[9] Due to the intrinsic features of the corresponding material, it has less than 200 wave number for conversion from photon to SPhP in the MIR to FIR region, which limits the operating frequency in a relatively narrow band. In contrast, SPP could be excited at a metal surface in principle in a broadband. However, the most suitable band for SPP is from visible to near-IR with acceptable mode confinement. The practical bandwidth is around 1 μm. Thus the suitable SPhP excitation bandwidth would be still comparable with that of SPP. Through introducing suitable superlattices, the negative dielectric response could be moved to different regions with engineerable bandwidth.[11,12]

As a derivation, when the thickness of the condensed matter becomes thinner enough, the SPhPs supported by the top and bottom surfaces will couple with each other to form a long range surface phonon polariton (LRSPhP) mode. The LRSPhP mode experiences lower propagation attenuation, which is desired in practical applications. As the attenuation is of most importance for such type of systems, we calculate the propagation length of LRSPhP modes for different thicknesses. The results are exhibited in Fig. 2. The mode is very lossy when the optical frequency is close to the resonance frequency. However, the loss of LRSPhP is also adjustable between the TO and LO frequencies. When the operation frequency is set away from resonance frequency, the propagation length could reach $10^3 \sim 10^4$ μm. Our following discussions are mainly focused on the LRSPhP mode.

As is mentioned above, the wavelength of SPhP is usually located in mid-infrared band. Due to this reason, the scale of an IR SPhP device may be 1-2 orders of magnitude larger than that of a typical visible SPP system. However, there are two major factors to determine the size of the electromagnetic surface mode, i.e., the frequency and the corresponding value of negative permittivity. For SPP modes operated at mid-infrared range, both the real and imaginary parts of the permittivity of metals are of 10^3 magnitudes. Thus the propagation constant would be pulled closer to the wave vector in the vacuum. Correspondingly, the SPP may have weaker confinement due to the too large negative dielectric constant. Although spoof SPPs have been recently proposed to increase the penetration of EM by cutting holes or grooves in metal surfaces. Complicated process has to be introduced. In contrast, for a SPhP system, the dielectric constant changes dramatically with in a relatively narrow

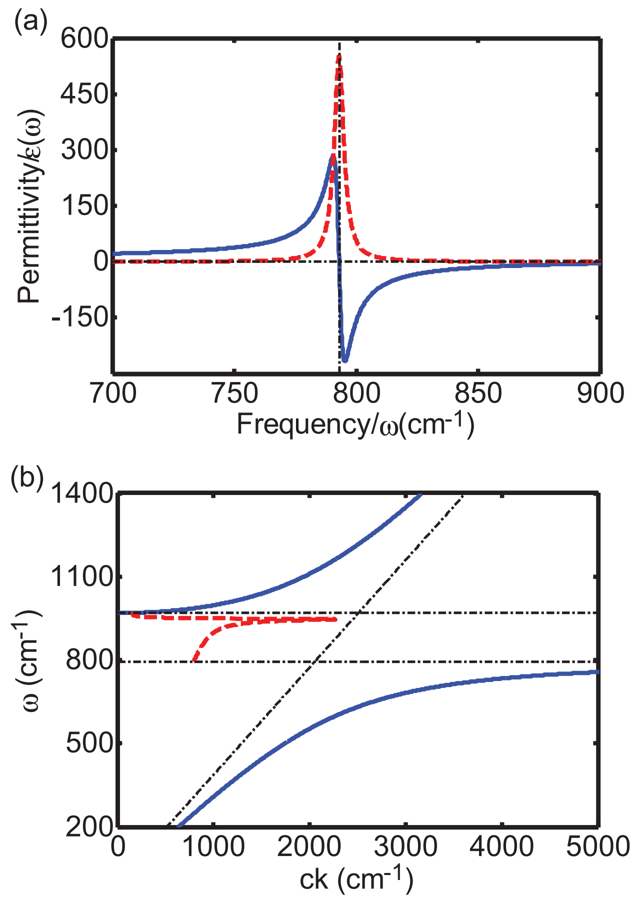

FIG. 1. (a) Dielectric permittivity curve of SiC, the blue solid line is the real part, while the red dashed line is the imaginary part; (b) Dispersion curve of SiC, the blue solid lines are those of bulk polaritons, while the red solid line is that of SPhP mode. The oblique dashed line is the light line.

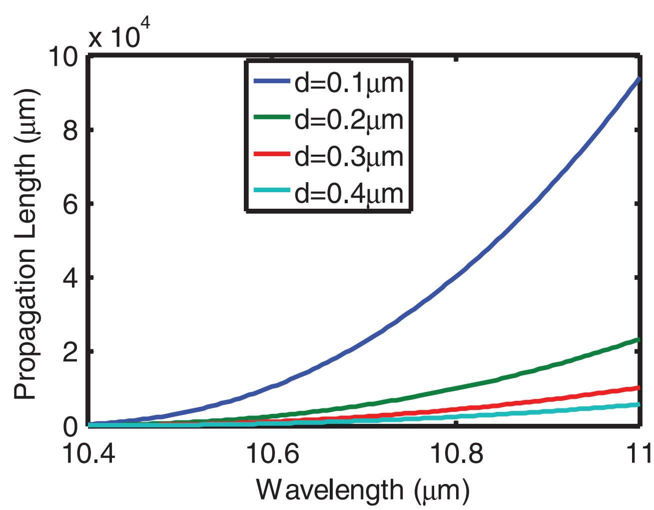

FIG. 2. Propagation length of LRSPhP modes corresponding to different thickness of the material strip.

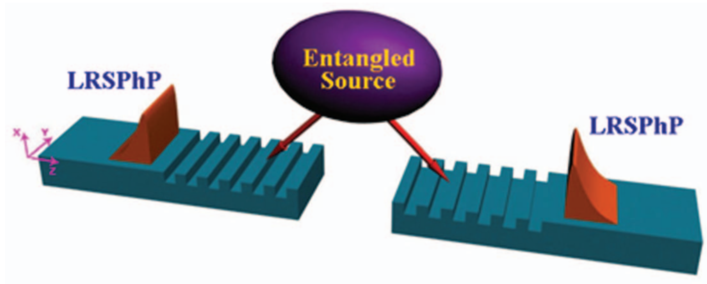

FIG. 3. Illustration of the generation of entangled LRSPhP modes.

range. In this case, smaller absolute value of negative dielectric constant gives smaller mode size and higher attenuation. On the contrary, larger value results in low propagation loss while maybe roomy just like the SPP. It gives the opportunity to select an operation frequency for a trade-off between mode size and propagation properties. This could be an advantage in designing SPhP based classical and quantum devices. From this point of view, SPhP may supply an alternative of sub-wavelength photonics in IR region.

The LRSPhP entangled pairs could be generated in the interaction process of entangled photons and the lossy condensed matter. For a clearer sense, an illustration of the model is presented in Fig. 3. When a pair of entangled photons incident on the condensed matters, they could be transferred into entangled SPhP pairs through surface gratings or the attenuated total reflection (ATR) regime. Although the usually used entangled photons sources are mostly in near-infrared band, nonlinear optical processes in crystals such as $ZnGeP_2$, CdSe and GaSe could be utilized to provide mid-infrared entangled photon pairs needed to generate entangled LRSPhP pairs.[13]

To describe the quantum characters and relative evolution processes of LRSPhP, a quantization formulation of LRSPhPs is of great importance. Ref. 14 gives an effective way to quantize the LRSPP mode through a canonical quantization procedure. That is of great value to study the lossy waveguide systems. Based on this way, the quantization of LRSPhP is obtained and the interaction Hamiltonian is derived. Further derivation through the perturbation theory could distinctly reveal the origin of the LRSPhPs entanglement. The quantization formulation of the vector potential could be expressed as

$$A(r,t) = \sum_k \sqrt{\frac{\hbar}{2\omega_k \varepsilon_0 L^2 |\Gamma_k|}}[b_k \Psi_k(x)\exp(i\beta_k z - i\omega_k t) + b_k^\dagger \Psi_k^*(x)\exp(-i\beta_k z + i\omega_k t)], \quad (3)$$

where the eigenvector $\Psi_k(r)$ could be expressed as

$$\Psi_k(x) = \left\{\left(\frac{i\beta_k}{\alpha_d}\hat{x} - \hat{z}\right)\exp[\alpha_d(x+h)]\theta(-x-h) + \left[\left(\frac{i\beta_k}{\alpha_c}\cdot\frac{\varepsilon_d\alpha_c}{\varepsilon_c\alpha_d}\hat{x} + \hat{z}\right)\cosh(\alpha_c x)\right.\right.$$
$$\left.\left. - \left(\frac{i\beta_k}{\alpha_c}\hat{x} + \frac{\varepsilon_d\alpha_c}{\varepsilon_c\alpha_d}\hat{z}\right)\sinh(\alpha_c x)\right]\theta(-x)\theta(x+h) + \left(\frac{i\beta_k}{\alpha_d}\hat{x} + \hat{z}\right)\exp(-\alpha_d x)\theta(x)\right\}. \quad (4)$$

The subscript is marked only by wave number k. b_k represents the annihilation operator of LRSPhP. α_q is given by $\alpha_q = \left(\beta_k^2 - \frac{\omega_k^2}{c^2}\varepsilon_q\right)^{\frac{1}{2}}$, $(q = d, c)$. Γ_k is the normalization coefficient which could be determined through the normalized longitudinal power flux.[15] The corresponding equation is written as $P_k = \frac{\beta_k}{2\omega_k\varepsilon_0|\Gamma_k|^2}\int_{-\infty}^{\infty}\frac{1}{\varepsilon_k(x)}|\Psi_k(x)|^2 dx = 1$.

Taken the quantization formulation of the LRSPhP mode, we could obtain the effective interaction Hamiltonian of the coupling process between entangled photons and entangled LRSPhPs. The classical Hamiltonian of the electromagnetic field is written as $H = \frac{1}{2}\int\limits_V\left(\varepsilon_0\varepsilon(x)E^2 + \mu_0 H^2\right)dv$. To describe the interaction process, the total electric field E_{tot} in the equation should be expressed as $E_{tot} = E_{eph} + E_{esp}$, in which E_{eph} and E_{esp} represents the field of entangled optical states and the field of entangled LRSPhPs, respectively. The optical field of entangled photons usually is generated by spontaneous parametric down-conversion (SPDC),[16] so the expression should be obtained from the second order nonlinear polarization $P = \varepsilon_0 \chi^{(2)} : EE$. Taking the actual interaction process into account, the electric field in the expression could be substituted by the $E^{(-)}$ part which is marked by the creation operator of photon $a_k^\dagger$ representing the producing of a photon.[1] For the entangled LRSPhPs, although the formulation of the field could not be derived through the second order nonlinearity, it is natural to determine that based on the correspondence between frequencies of the incident photons and the LRSPhP modes. Substituting the relevant formulations into the Hamiltonian of the electromagnetic field, the overall interaction Hamiltonian is obtained as follows

$$H = \frac{\pi\hbar^2(\chi^{(2)})^2}{2\varepsilon_0} \sum_k \sum_{(\sigma_1,\sigma_2)} I_{k\sigma_1\sigma_2} a^\dagger_{k\sigma_1} a^\dagger_{(k_p-k)\sigma_2} a_{(k_p-k)\sigma_2} a_{k\sigma_1} + \frac{\pi\hbar^2}{2\varepsilon_0 L^4} \sum_{k_1,k_2} I_{k_1k_2} b^\dagger_{k_1} b^\dagger_{k_2} b_{k_2} b_{k_1}$$
$$+ \left\{ \sum_{k,k_1,k_2} \sum_{(\sigma_1,\sigma_2)} \Im_{kk_1k_2} \mathbb{C}(r_1, r_2) b^\dagger_{k_1} b^\dagger_{k_2} a_{k\sigma_1} a_{(k_p-k)\sigma_2} \exp[-i(\omega_p - \omega_{k_1} - \omega_{k_2})t] + H.C. \right\}, \quad (5)$$

where

$$I_{k\sigma_1\sigma_2} = \frac{\omega_{k\sigma}(\omega_p - \omega_{k\sigma})}{\Xi_1 \Xi_2} \int dx_1 dx_2 \varepsilon(x_1)\varepsilon(x_2) \left|\Theta(x_1, x_2)\right|^2$$
$$I_{k_1k_2} = \frac{\omega_{k_1}\omega_{k_2}}{\Gamma_{k_1}\Gamma_{k_2}} \int dx_1 dx_2 \varepsilon(x_1)\varepsilon(x_2) \left|\Psi_{k_1}(x_1)\right|^2 \left|\Psi_{k_2}(x_1)\right|^2 .$$

The quantities marked by subscript p are relative parameters of the pump field of the SPDC process, and the perfect phase matching conditions are assumed to be satisfied. Ξ_1 and Ξ_2 are defined by respective normalization procedures. The eigenvector $\Theta(x_1, x_2)$ could be decomposed into the product of eigenvectors of the two subspaces $\Theta(x_1, x_2) = \vartheta^*(x_1)\vartheta^*(x_2)$. The detailed formulation of $\vartheta(x)$ could be expressed as[14]

$$\vartheta(x) = [\lambda_1 \exp(-\alpha_c x) + \lambda_2 \exp(\alpha_c x)]\theta(-x)\theta(x+h) + \lambda_3 \exp[\alpha_d(x+h)]\theta(-x-h), \quad (6)$$

where the λ_i's (i = 1, 2, 3) are determined by the overall boundary conditions of the coupling gratings.

In the interaction Hamiltonian, the first and the second terms corresponds to the unperturbed portion of the Hamiltonian. The physical meaning of these two terms is quite clear. Their effective operators are both combinations of the particle number operators for the entangled states, so expectation values of these operators represent the combination detections of entangled photons and entangled LRSPhPs, respectively. The latter two terms corresponds to the interaction of entangled states. The first one describes the generation of the entangled LRSPhPs, while the latter one represents the conjugate process. So the effective H_1 could be written as

$$H_1 = \sum_{k,k_1,k_2} \sum_{(\sigma_1,\sigma_2)} \Im_{kk_1k_2} \mathbb{C}(r_1, r_2) b^\dagger_{k_1} b^\dagger_{k_2} a_{k\sigma_1} a_{(k_p-k)\sigma_2} \exp[-i(\omega_p - \omega_{k_1} - \omega_{k_2})t]. \quad (7)$$

Here the coefficient $\Im_{kk_1k_2}$ is given by

$$\Im_{kk_1k_2} = \frac{\hbar^2}{4\varepsilon_0 L^2} \sqrt{\frac{\omega_{k_1}\omega_{k_2}}{\Gamma_{k_1}\Gamma_{k_2}}} \sqrt{\frac{\omega_{k\sigma}(\omega_p - \omega_{k\sigma})}{\Xi_1 \Xi_2}}. \quad (8)$$

In the derivation, we assume the conservation of energy is satisfied in the SPDC process to prepare the entangled optical fields, so the spatial integral in the interaction Hamiltonian is estimated as

$$\mathbb{C}(r_1, r_2) = \int d^3 r_1 \int d^3 r_2 h(r_1, r_2)$$
$$= \left(\int dx_1 dx_2 \varepsilon(x_1)\varepsilon(x_2) \Psi^*_{k_1}(x_1) \Psi^*_{k_2}(x_2) \Theta^*(x_1, x_2) \right) \cdot \sin c \left(\frac{\Delta\beta_1 z_1}{2} \right) \sin c \left(\frac{\Delta\beta_2 z_2}{2} \right) z_1 z_2. \quad (9)$$

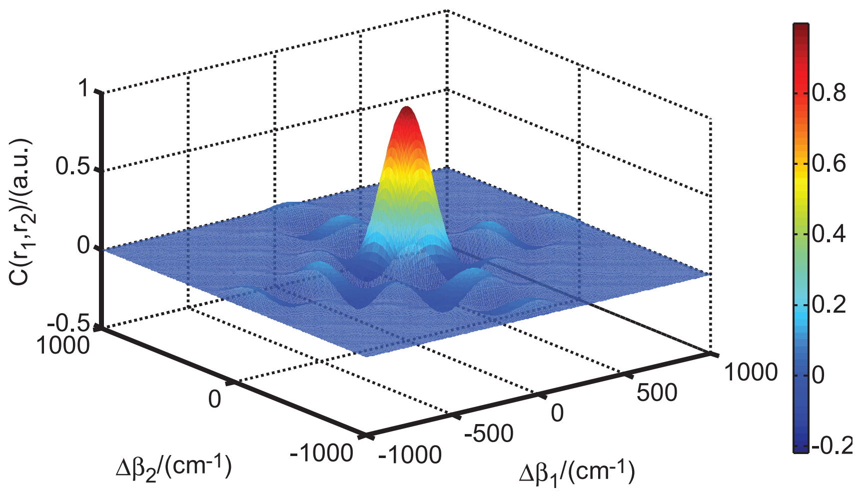

FIG. 4. Normalized profile of $\mathbb{C}(r_1,r_2)$. Both z_1 and z_2 are set at 100 μm.

To simplify the form of expression, origins of z_1 and z_2 axes are set in the middle of the gratings, *i.e.*, the coupling sections. $\Delta\beta_1$ and $\Delta\beta_2$ are the relative wave vector mismatches of the coupling processes, $\mathbb{C}(r_1, r_2)$ is plotted as a function of these two quantities as shown in Fig. 4. In the calculations, z_1 and z_2 are both set at 100 μm.

For a more explicit comprehension of the entangled LRSPhP modes, the state vector could be derived from second order perturbation

$$|\Psi\rangle = \left(-\frac{i}{\hbar}\right)^2 \int dt_1 \int dt_2 T[H_{SPDC}(t_1)H_1(t_2)]\,|0\rangle, \tag{10}$$

where T is the time-ordering operator. Neglecting the unrelated items, $|\Psi\rangle$ could be written as

$$|\Psi\rangle = -\frac{2\pi}{\hbar^2}\sum_{k_1,k_2} F_k \Im_{kk_1k_2}\mathbb{C}(r_1, r_2)\delta(\omega_p - \omega_{k_1} - \omega_{k_2})b^\dagger_{k_1} b^\dagger_{k_2}\,|0\rangle\,. \tag{11}$$

The coefficient F_k is defined by the SPDC process. We would like to take the entangled state for a detailed consideration. As we know, the SPDC-generated entangled photon state is usually entangled in frequency and wave vector.[1] In frequency space, the coupling process from photon to LRSPhP does not influence the corresponding frequency property, so the frequency entanglement derived from entangled photons could completely preserve in the entangled LRSPhPs, which is stressed by the Dirac-δ function in Eq. (11). For the case of wave vector, the situation is slightly different. The wave-vectors along z direction should be considered for the phase-matching condition of the coupling process. Generally speaking, the wave-vector entanglement has implications for the spatial correlations of the entangled pair,[1] so the quantum entanglement based on SPhP corresponds to a low dimension spatial correlation system. In order to quantify the entanglement, we calculate the concurrence of LRSPhP entangled state based on the SiC system. The concurrence $C(\Psi)$ is defined to be $C(\Psi) = \frac{|\langle\Psi|(\sigma_y\otimes\sigma_y)|\Psi^*\rangle|^2}{|\langle\Psi|\Psi\rangle|^2}$, where σ_y is the Pauli operator $\begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix}$. Substituting Eq. (11) into the definition, we have

$$\begin{aligned} C(\Psi) &= \frac{4\pi^2}{\hbar^4} \sum_{k_1,k_2,k_1',k_2'} \frac{F_k^2 \Im_{kk_1k_2}^2 \left|\mathbb{C}(r_1,r_2)\right|^2}{\left|\langle\Psi|\Psi\rangle\right|^2} \delta(\omega_p - \omega_{k_1} - \omega_{k_2}) \\ &\quad \times \delta(\omega_p - \omega_{k_1}' - \omega_{k_2}') \left\langle 0\right| b_{k_2}^{\dagger} b_{k_1}^{\dagger} (\sigma_y \otimes \sigma_y) b_{k_1'} b_{k_2'} \left|0\right\rangle \\ &= \frac{4\pi^2}{\hbar^4 \left|\langle\Psi|\Psi\rangle\right|^2} \sum_{k_1,k_2,k_1',k_2'} F_k^2 \Im_{kk_1k_2}^2 \left|\mathbb{C}(r_1,r_2)\right|^2 \delta(\omega_p - \omega_{k_1} - \omega_{k_2}) \\ &\quad \times \delta(\omega_p - \omega_{k_1}' - \omega_{k_2}') \left\langle 0\right| (b_{k_1}^{\dagger} b_{k_2'})_A (b_{k_2}^{\dagger} b_{k_1'})_B \left|0\right\rangle \end{aligned} \tag{12}$$

In this equation, subscript A and B correspond to two subspaces. When we have $k_1 = k_2'$ and $k_2 = k_1'$, the value of $C(\Psi)$ is 1, so the LRSPhP entangled state is of complete entanglement.

III. STATES TRANSFORMATION AND CORRELATIONS OF SYSTEM

For a further consideration, we notice that in the coupling process of entangled photons and entangled LRSPhPs, the former one could not been transformed into the latter one with unity probability. Referring to the classical situation, the coupling coefficient determines the portion of the incident light which could be coupled into LRSPhP mode. For the quantum situation, that would correspond to the transition probability from a photon to an LRSPhP. Therefore there are in fact three possible results of the interaction between entangled photons and the grating structures. Besides the transformation from entangled photons to entangled LRSPhPs we have discussed above, it still may generate an entangled pair of photon and LRSPhP, or a surviving entangled photon pair. For a treatment of the state transformation processes, if the quantum field theory (QFT) method is used, there will be many effective terms of the total interaction Hamiltonian to be treated. Thus the obtained state vector is in relatively complex formation. Therefore we may use the wave mechanics (WM) method as a substitution. This method is based on the Bialynicki-Birula-Sipe equation,[17,18] which has a similar formation with the Schrödinger equation for electron. A photon is described by the so-called "photon wave function." The equation is proved to be equivalent to the set of Maxwell equations,[19] which makes it seem classical. However, it has been verified that the WM theories of photon could be interchangeable with the QFT theories. Through the WM method, we obtain the transformation matrix directly, which gives an intuitive description of the coupling process.

To investigate variations of two-entity states, the Bialynicki-Birula-Sipe equation should be generalized. The motion equation for the two-photon wave function in vacuum has been derived by Smith and Raymer.[19,20] We need to expand the equation in dealing with problems in dielectrics. The Bialynicki-Birula-Sipe equation for single photon wave function in dielectrics has been derived in Ref. 21, through a similar treatment; the governing equation for two-state evolution is obtained as

$$i\frac{\partial}{\partial t}\Psi_d^{(2)} = c\alpha_1^{(2)}\nabla_1 \times \Psi_d^{(2)} + c\alpha_2^{(2)}\nabla_2 \times \Psi_d^{(2)}, \tag{13}$$

where

$$\alpha_1^{(2)} = \Sigma_3 \otimes I, \alpha_2^{(2)} = I \otimes \Sigma_3,$$

with

$$I = \begin{bmatrix} \hat{1} & 0 \\ 0 & \hat{1} \end{bmatrix},\ \Sigma_3 = \begin{bmatrix} \hat{1} & 0 \\ 0 & -\hat{1} \end{bmatrix},\ \hat{1} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}.$$

The two-state wave function $\Psi_d^{(2)}$ is written as

$$\Psi_d^{(2)}(r_1, r_2, t) = \sum_{(j\sigma),(m\rho)} C_{(j\sigma),(m\rho)} \psi_{j\sigma}^{(1)}(r_1, t) \otimes \psi_{m\rho}^{(1)}(r_2, t). \tag{14}$$

Here (jσ) and (mρ) are subscripts of state 1 and state 2, respectively. The single state wave function in dielectrics is written as

$$\begin{gathered}\psi^{(1)}(r,t)=\psi_{+}^{(1)}(r,t)+\psi_{-}^{(1)}(r,t).\\ \left(\psi_{\pm}^{(1)}(r,t)=\sqrt{\frac{\varepsilon_0\varepsilon}{2}}E_{\pm}\pm i\sqrt{\frac{\mu_0}{2}}H_{\pm}\text{ and }\hat{\sigma}\psi_{\pm}^{(1)}=\pm\psi_{\pm}^{(1)}\right)\end{gathered}\tag{15}$$

The actual electric field and magnetic field of the system are $E=E_{+}+E_{-}$ and $H=H_{+}+H_{-}$, respectively. It should be emphasized that the photon wave function could not be normalized in the usual sense based on position probability density due to the lack of localizability.[19] As the energy density of the electromagnetic field is localized, it is applied as a substitution in WM of photon, which is equal to the moduli square of Ψ(r, t). For the LRSPhP mode, as the energy distribution is inhomogeneous due to the space dependence of permittivity, the corresponding wave function of LRSPhP whose moduli square represents the spacial energy density perhaps possesses potential value to describe the quantum behavior of LRSPhP.

It is noteworthy that these equations are only reasonable for non-absorptive matters. Although the real systems we discussed are lossy, the key process is still the coupling between entangled photons and LRSPhP modes. The propagation attenuation doesn't play a crucial role, thus the imaginary part of the lossy material could be ignored in the derivation. Based on the theories above, the controlling equation of the interaction process is given by

$$\sum_{\nu,\eta}\exp[-i(\Delta\beta_{1,\nu\mu}z_1+\Delta\beta_{2,\eta\tau}z_2)]F_{\nu\mu}(r_1)F_{\eta\tau}(r_2)=0.\tag{16}$$

In the equation, the function $F_{\nu\mu}(r)$ is written as

$$F_{\nu\mu}(r)=\left(\frac{\partial}{\partial z}-i\Delta\beta_{\nu\mu}\right)\sum_{(\nu,\mu)}\operatorname{Im}\langle\mathrm{Re}\psi_\nu|\frac{\hat{A}}{\sqrt{\varepsilon(x)}}\left|\psi_\mu\right\rangle_S-\frac{i\omega}{c\varepsilon_0}\langle\mathrm{Re}\psi_\nu|\frac{\Delta\varepsilon}{\varepsilon(x)}\left|\mathrm{Re}\psi_\mu\right\rangle_S,\tag{17}$$

where the operator $\hat{A}$ is equivalent to $\times\,\hat{\sigma}$. And, the function $F_{\eta\tau}(r)$ has a fully corresponding formulation. Through further derivations, the equation could be simplified to

$$\sum_{\nu,\eta}\exp[-i(\Delta\beta_{1,\nu\mu}z_1+\Delta\beta_{2,\eta\tau}z_2)]\partial'_{z_1}\partial'_{z_2}\xi^{(2)}_{\nu\eta}(z_1,z_2)=0.\tag{18}$$

The $\xi^{(2)}_{\nu\eta}(z_1,z_2)$ represents the combination of the longitudinal portion of the wave functions, and the operator ∂'_{z_j} is expressed as

$$\partial'_{z_j}=\lambda_{(\nu\mu),(\eta\tau)}\left(\frac{\partial}{\partial z_j}-i\Delta\beta_{(\nu\mu),(\eta\tau)}\right)+i\omega_j\kappa_{(\nu\mu),(\eta\tau)}.\tag{19}$$

$\lambda_{(\nu\mu),(\eta\tau)}$ and $\kappa_{(\nu\mu),(\eta\tau)}$ represents the orthonormalization and coupling coefficients, respectively. Through these functions, the states transformation process is expressed as

$$\begin{bmatrix}\xi_{\nu\eta,f}\\ \xi_{\nu\tau,f}\\ \xi_{\mu\eta,f}\\ \xi_{\mu\tau,f}\end{bmatrix}=\begin{bmatrix}t_{\nu\mu}t_{\eta\tau} & t_{\nu\mu}\kappa_{\eta\tau} & \kappa_{\nu\mu}t_{\eta\tau} & \kappa_{\nu\mu}\kappa_{\eta\tau}\\ -t_{\nu\mu}\kappa^*_{\eta\tau} & t_{\nu\mu}t^*_{\eta\tau} & -\kappa_{\nu\mu}\kappa^*_{\eta\tau} & \kappa_{\nu\mu}t^*_{\eta\tau}\\ -\kappa^*_{\nu\mu}t_{\eta\tau} & -\kappa^*_{\nu\mu}\kappa_{\eta\tau} & t^*_{\nu\mu}t_{\eta\tau} & t^*_{\nu\mu}\kappa_{\eta\tau}\\ \kappa^*_{\nu\mu}\kappa^*_{\eta\tau} & -\kappa^*_{\nu\mu}t^*_{\eta\tau} & -t^*_{\nu\mu}\kappa^*_{\eta\tau} & t^*_{\nu\mu}t^*_{\eta\tau}\end{bmatrix}\begin{bmatrix}\xi_{\nu\eta,i}\\ \xi_{\nu\tau,i}\\ \xi_{\mu\eta,i}\\ \xi_{\mu\tau,i}\end{bmatrix}.\tag{20}$$

In our analysis, the only initial state is the entangled photon state. Through the transformation, four entangled states in three types are generated with corresponding possibilities.

For a clear view, a practical instance of coupling is given in Fig. 5. It is based on SiC systems. The incident angle is set at 6°, and the grating period is chosen to be 13 μm. The coupling efficiencies for different parameters are obtained indirectly through the rigorous coupled wave analysis (RCWA) method. From Fig. 5(a), at the excitation wavelength 11.9 μm, coupling efficiency κ stays above 0.6, which corresponds to the transformation efficiency from photon to LRSPhP. Thus the generation probability of the entangled LRSPhP pairs, entangled LRSPhP with photon and entangled photon pairs could be calculated correspondingly, which are equal to $|\kappa|^2$, $2\kappa(1-\kappa)$

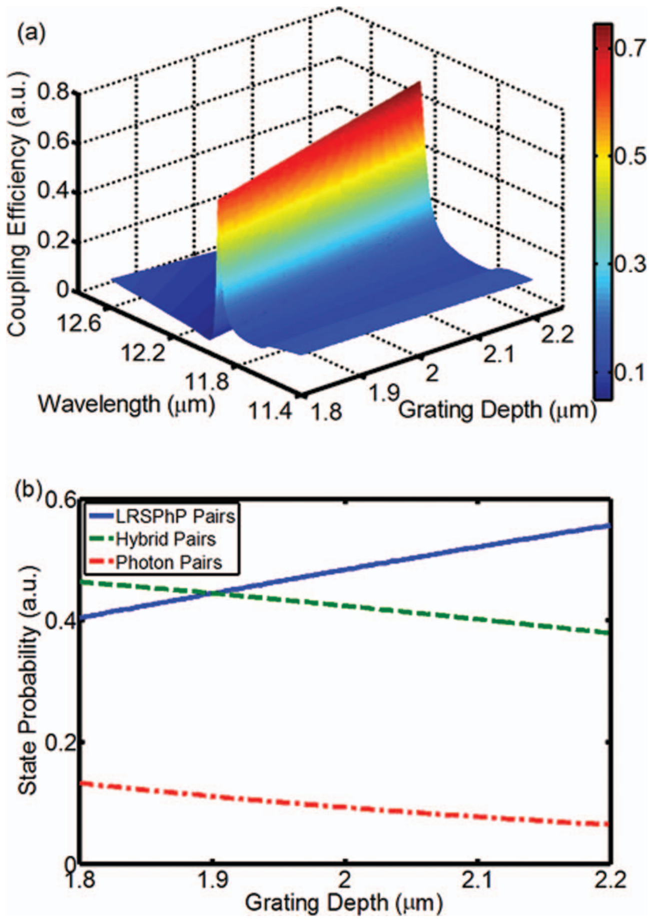

FIG. 5. (a) Coupling efficiency from photon to LRSPhP. The incident angle is 6°, and the grating period is 13 μm. The wavelength for effective coupling presents at 11.9 μm. The coupling efficiencies stay between 0.6 and 0.8. (b) Generation probability of three types of entangled states. The blue solid line corresponds to the entangled LRSPhP state. The hybrid entangled state of LRSPhP with photon is represented by the green dashed line, while the red dotted line is for the entangled photon state.

and $|(1-\kappa)|^2$ respectively. The state probabilities changing with the depth of grating are plotted in Fig. 5(b). The generation probabilities of the entangled LRSPhP state and the hybrid entangled state of LRSPhP with photon are around 0.45, while that of the entangled photon state is about 0.1.

To show correlations of the system, the second-order correlation function has been investigated. It could be derived from tracing over the tensor product of the two-photon wave function and its Hermitian conjugate then integrating over all space. The formulation of the correlation function could be written as

$$\gamma^{(2)}(r_1, r_2; t) = \int\int Tr[\Psi^*(r_1, r_2, t)\Psi(r_1, r_2, t)]d^3r_1 d^3r_2, \tag{21}$$

For an unentangled case, the numerator of the correlation function could be decomposed into products of the subspaces. In contrast, the correlation function for entangled states will include coherent superposition of the states which implies the nonlocal correlations. The correlation function is expressed as

$$\gamma^{(2)}(r_1, r_2; t) \propto F_{\alpha\beta}\, g(r_1, r_2; t), \tag{22}$$

where $F_{\alpha\beta}$ is determined by the specific entangled pair, and (α, β) represents photon or LRSPhP. For a clear view, we take the energy-time type as an illustration. The detailed formulation of the two-state wave function could be written as[20]

$$\Psi_d^{(2)}(r_1, r_2, t) = \sum_{jm} C_{jm} \left(\sqrt{\frac{\varepsilon_0 \varepsilon}{2}} E_{j,\pm} \pm i \sqrt{\frac{\mu_0}{2}} H_{j,\pm} \right) \otimes \left(\sqrt{\frac{\varepsilon_0 \varepsilon}{2}} E_{m,\pm} \pm i \sqrt{\frac{\mu_0}{2}} H_{m,\pm} \right), \tag{23}$$

with

$$E_{i,\pm} = E^0_{i,\pm}(t) + E^0_{i,\pm}(t - T_i).$$

T_i ($i = 1, 2$) is the time delay introduced in by the Franson interferometer. The magnetic field has the similar formulation. Correspondingly, the general expression is given as follows

$$\begin{aligned} &g(t) \propto \sum_i |G(t_i)|^2 + \sum_{i \neq j} G(t_i) G^*(t_j) \\ &t_i, t_j \in \{(t, t), (t - T_1, t), (t, t - T_2), (t - T_1, t - T_2)\}, \end{aligned} \tag{24}$$

with

$$G(t_1, t_2) \propto \int\int d\omega_1 d\omega_2 \Phi(\omega_1, \omega_2) e^{-[\mathrm{Im}(\beta_1(\omega_1))L_1 + \mathrm{Im}(\beta_2(\omega_2))L_2]} e^{-i\omega_1 \cdot t_1 - i\omega_2 \cdot (t_2 + \tau)}.$$

In this equation, $\Phi(\omega_1, \omega_2)$ is generated from the SPDC process. The item $e^{-[\mathrm{Im}(\beta_1(\omega_1))L_1 + \mathrm{Im}(\beta_2(\omega_2))L_2]}$ is determined by the intrinsic propagation loss of the LRSPhP. Referring to Ref. 4, for the energy-time systems, that would cause a certain extent of loss. The received signals correspondingly experience decays. Fortunately, it does not affect the degree of entanglement. The second item represents the contribution from the nonclassical correlation of the system. In the actual situation, we usually have T_1, $T_2 \gg T_c$ (the coincidence window), so the effective terms are dependent on (t, t) and ($t - T_1$, $t - T_2$). Thus Eq. (24) is simplified as follows

$$g(t) \propto |G(t, t)|^2 + |G(t - T_1, t - T_2)|^2 + \left[G(t, t) G^*(t - T_1, t - T_2) + c.\,c. \right] \tag{25}$$

with

$$|G(t_1, t_2)|^2 = \int\int d\omega_1 d\omega_2 \int\int d\omega_1' d\omega_2' V(\omega_1, \omega_2) V^*(\omega_1', \omega_2') e^{-i(\omega_1 - \omega_1')\cdot t_1 - i(\omega_2 - \omega_2')\cdot(t_2 + \tau)}$$

$$\begin{aligned} G(t, t) G^*(t - T_1, t - T_2) = &\int\int d\omega_1 d\omega_2 \int\int d\omega_1' d\omega_2' V(\omega_1, \omega_2) \\ &\times V^*(\omega_1', \omega_2') e^{-i(\omega_1 - \omega_1')\cdot t - i(\omega_2 - \omega_2')\cdot(t + \tau) - i(\omega_1' T_1 + \omega_2' T_2)} \end{aligned}$$

In the equation above, we have set $V(\omega_1, \omega_2) = \Phi(\omega_1, \omega_2) e^{-[\mathrm{Im}(\beta_1(\omega_1))L_1 + \mathrm{Im}(\beta_2(\omega_2))L_2]}$. After time integration and normalization, the function could be expressed as

$$\begin{aligned} \Gamma^{(2)} &\propto \frac{\int\int d\omega_1 d\omega_2 \, |V(\omega_1, \omega_2)|^2 \left[2 + e^{-i(\omega_1 T_1 + \omega_2 T_2)} + e^{i(\omega_1 T_1 + \omega_2 T_2)} \right]}{\int\int d\omega_1 d\omega_2 \, |V(\omega_1, \omega_2)|^2} \\ &\propto \frac{\int\int d\omega_1 d\omega_2 \, |V(\omega_1, \omega_2)|^2 \left[1 + \cos(\omega_1 T_1 + \omega_2 T_2) \right]}{\int\int d\omega_1 d\omega_2 \, |V(\omega_1, \omega_2)|^2} \end{aligned} \tag{26}$$

For the case of perfect phase matching, the formation could be simplified into a cosine function. The corresponding results are shown in Fig. 6. The visibility is sure to be unity, which verifies the robust nonlocality correlation in theory.

IV. CONCLUSIONS

In summary, we investigate the entanglement of LRSPhP modes in condensed matter systems. Comparing with the previously reported quantization theory of LRSPP,[14] the differences are shown in some aspects. For example, LRSPhP is a phonon mode reflecting the collective excitations of ions in polar dielectric materials while the SPPs exhibit in dielectric/metallic systems. Due to the longer resonance wavelength, the SPhP systems normally work in the mid-infrared band, while the ordinary

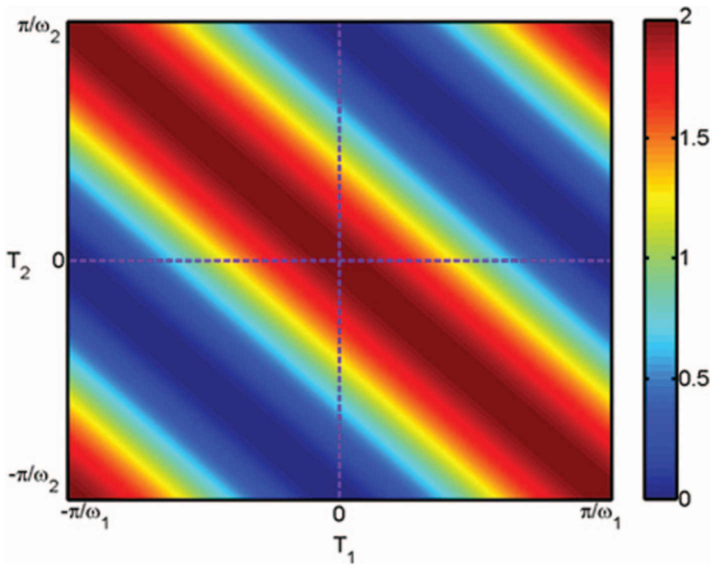

FIG. 6. Values of function $\Gamma^{(2)}$ for the case of perfect phase matching.

operation wavelengths of SPP are in visible to near-infrared range. In the derivation process of Ref. 14, density matrix theory is employed to describe coherent LRSPP states. However, in our study, the interaction Hamiltonian is derived and the perturbation theory is utilized to obtain the state vector of the LRSPhP entangled pairs. The origin of LRSPhP entanglement thus is revealed. Moreover, we attempt to use the wave mechanics approach alternatively in describing the coupling process. The correlation of system is also illustrated based on the energy-time arrangement. The entanglement of LRSPhP indirectly verifies the single quantum property of the "collective excitation". Since LRSPhP has unique capability to confine the electromagnetic field in the perpendicular direction, it should be a promising candidate for integrated quantum systems in mid-infrared band.

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