We explore a mechanism for producing time-frequency entangled photon pairs (termed a biphoton) from an ensemble of atom-like solid-state quantum emitters. Four distinct energy levels of the solid-state system render four spin-conserving optical transitions as observed in color centers. This feature opens up the possibility to generate a four-wave mixing biphoton based on an electromagnetic induced transparency (EIT) for long-coherence quantum communication as demonstrated in cold atomic systems. We propose a narrow EIT window below a lifetime-limited linewidth of a SiV\textsuperscript{−} in diamond, assuming a few hundred MHz. Consequently, the EIT-induced narrowband guarantees biphoton coherence time to be at least a few tens of a nanosecond without a cavity. Assessing the criteria of solid-state parameters applicable to the existing biphoton model from cold atoms will accelerate solid-state biphoton source research. This study shows that a realization of negligible ground state dephasing of a solid-state sample will be a crucial step toward a solid-state biphoton generation for more than a 100 ns time scale with a subnatural atomic linewidth of a few MHz.

A narrowband biphoton source is an essential ingredient of quantum communications, especially for long-coherence quantum networks based on quantum entanglement [1,2]. A conventional protocol [3] has been tested based on spontaneous parametric down conversion (SPDC) using a nonlinear crystal [4,5]. For practical implementation, the most challenging issue of the conventional biphoton generation method has been short coherence time less than a pico-second (THz) due to the broadband linewidth of nonlinear crystals. The coherence timescale should be at least more than several nanoseconds, which are longer than detection time resolution. For such a timescale up to tens of nanoseconds, the SPDC nonlinear crystal is located inside an optical cavity, and the paired bandwidth has been decreased by 10 MHz [6–13].

A method to control biphoton pairs could be robust if we further increase the coherence timescale. A longer timescale, for example, secures efficient interaction between the heralded single photon and atomic ensemble, which can engineer an entanglement in a temporally correlated scheme [14]. Recently, easily tunable long-coherence time from 50 to 900 ns has been demonstrated as the first subnatural narrowband entangled photon pair generation by utilizing electromagnetically induced transparency (EIT)-based spontaneous four-wave mixing (SFWM) in a laser-cooled cold atomic ensemble [15]. Compared to the cavity-based narrowband SPDC, the EIT-based SFWM cold atomic technique [15–19] has been developed so that the timescale was increased up to a microsecond order [16]. The longer coherence time compared to detection time is the actual benefit for obtaining a high purity of a single photon [20] if we consider about a nanosecond single photon detection time. However, most of these previous EIT-based SFWM narrowband biphoton studies were done in a magneto-optical trap, which requires bulky, complicated apparatus. For this process to be implemented in a real application, we need miniaturization and the capability for photonic chip integration. A hot atomic biphoton source [21,22] would be the intermediate step for the miniaturization. Up to now, there have been several trials to demonstrate fundamental quantum optics in atom-like solid-state systems, such as rare-earth-doped material [23], a quantum dot [24], a nitrogen-vacancy center in diamond [25,26], or a negatively charged silicon-vacancy center in diamond (SiV\textsuperscript{−}) [27,28]. Most recently, researchers demonstrated a bright single-photon source using SiV\textsuperscript{−} embedded nanowire array [29]. The nanowire array might improve light
extraction efficiency, so that the integration of a solid-state quantum light source becomes reliable. However, there has been no research activity toward replacing the atomic ensemble with a new solid-state material for the narrowband biphoto generation.

In this article, we assess criteria to determine parameters for the narrowband biphoto generation from atom-like solid-state emitters, such as quantum dots, nitrogen-vacancy or silicon-vacancy centers in diamond via the EIT-SFWM process. Here, we find a parameter regime to apply the existing biphoto model from a cold atomic ensemble to our proposed solid-state system instead of building a new model that only applies to a solid-state system. The idea is to use an atom-like spectrum of solid-state material, for example, negatively charged silicon vacancy in diamond defect centers (SiV−). The spectral properties of SiV− show strong optical transition due to a zero-phonon line (ZPL) near 737 nm with weak phonon sidebands [30]. This approach will be used to realize a bandwidth that is even narrower than the lifetime-limited linewidth of the solid-state sample (a few hundred MHz), as has been previously accomplished in an atomic ensemble by achieving less than a natural linewidth (6 MHz). Although further research for achieving a few MHz narrow linewidths like atomic transitions rather than the currently demonstrated 100 MHz is a requirement in material science, our proposed study will pave the way for the implementation of a narrowband biphoto generator towards silicon-based integrated photonic chips for quantum information processing.

The criteria of biphoto generation in solid-state platforms come from spectra of allowed energy levels, linewidths, dipole moments, and polarizations. Our proposed energy scheme considers a double Λ configuration [15] consisting of four energy levels, two excited states, and two ground states, associated with four peaks in absorption spectrum. Each energy level consists of two degenerate levels under zero B-field [28,30]. The optical transition properties of SiV− are relevant to low energy gaps that originate from carbon dangling bonds [30].

Figure 1 compares experimental schemes and energy levels of: (a) a cold rubidium (Rb) atomic ensemble [15], and (b) a proposed SiV− diamond case. For both cases, a counter-propagating pump (736.4 nm, \(\omega_p\)) and coupling (737 nm, \(\omega_s\)) fields shining on the sample spontaneously generate Stokes (\(\omega_s\)) and anti-Stokes (\(\omega_a\)) photon pairs by SFWM in the double Λ energy configuration consisting of an off-resonance Raman scattering and EIT. Differences between the two systems are shown in Figs. 1(a) and 1(b). Compared to 15 nm splitting between excited states in Fig. 1(a), proposed excited states are separated by 0.5 nm in wavelength or 258 GHz in frequency, still providing enough spectral resolution between two Λ configurations. Compared to 3.035 GHz ground splitting in \(^{87}\)Rb or 6.834 GHz in \(^{85}\)Rb atoms, the wide ground state splitting, 46 GHz [28,30], of our proposed scheme is meritorious regarding noise reduction in the biphoto generation.

Broadening of a linewidth \(\Gamma_0\) is the crucial factor in determining coherence time for biphoto generation. The narrowest linewidth of SiV− diamond defect centers is less than 100 MHz, 94 MHz, which is almost limited by its lifetime, 1.7 ns [28]. In general, however, SiV− diamond has about a GHz (FWHM) broadening even at cryogenic temperatures [30]. So, our model considers a 160 MHz broadened line by assuming 1 ns lifetime at low temperature of 4 K, and our goal is to achieve less than 100 MHz biphoto linewidth compared to our proposed 160 MHz in this paper. Note that the subnatural linewidth demonstrated in cold atoms [15] is a linewidth...
narrower than the atomic natural linewidth, 6 MHz. This approach will be directly used, if applicable, for the case of a few MHz narrowband atomic-like solid-state samples when we overcome broadening issues in the future. In contrast to inhomogeneous broadening of atoms depending on temperature (Doppler broadening), spectral broadening in solid-state systems occurs for various reasons: temperature (phonon), crystal symmetry, and strain-induced broadening [31]. Despite the intrinsic strain-induced broadening of heavy atomic defects like N or Si, defect centers show a sharp ZPL. To further reduce the broadening, we consider SiV− rather than NV in our model because of its $D_{4h}$ crystal structural symmetry [30].

While there are theoretical studies of electronic structure of SiV− defects using ab initio [32], explicit values of dipole moments are not available yet. So, we may extract such values from a measured absorption spectrum. For the case of SiV−, an optical dipole transition is only allowed for a spin conserving relaxation from excited states to ground states [30].

An empirically determined polarization of each optical transition is characterized by a crystal axis and a converted frame of reference (Doppler broadening), spectral broadening in solid-state systems occurs for various reasons: temperature (phonon), crystal symmetry, and strain-induced broadening [31]. Despite the intrinsic strain-induced broadening of heavy atomic defects like N or Si, defect centers show a sharp ZPL. To further reduce the broadening, we consider SiV− rather than NV in our model because of its $D_{4h}$ crystal structural symmetry [30].

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We apply a biphoton model of an atomic system [33] to the SiV− case. Under the condition of energy consideration, we consider the entangled state given by

$$|\Psi\rangle = L \int d\omega \kappa(\omega_{\alpha}, \omega_{\beta}) \text{sgn}(\Delta k) \left( a_{\alpha}^\dagger(\omega_{\alpha}) a_{\beta}^\dagger(\omega_{\beta}) |0\rangle \right),$$

where $L$ is medium length, and $\kappa(\omega_{\alpha}, \omega_{\beta}) = -i(\sqrt{\omega_{\alpha} \omega_{\beta}} / 2e) \chi^{(3)}(\omega_{\alpha}, \omega_{\beta}) E_{\alpha} E_{\beta}$ is a nonlinear coupling coefficient. Here, $E_{\alpha}, E_{\beta}$ is pump (coupling) light amplitude. $\Delta k = (\vec{k}_{\alpha} - \vec{k}_{\beta}) \cdot \vec{k}$ describes a phase matching condition and a third-order nonlinear susceptibility related to the four-wave mixing is given by

$$\chi^{(3)}(\omega_{\alpha}, \omega_{\beta}) = \frac{N \mu_1 \mu_2 \mu_3 \mu_4 \mu_{12}}{4 \epsilon_0 h^3 (\Delta_{\omega}^\gamma + i |\gamma|) (\omega^2 - (\Omega^2 / 2)^2)^2}.$$

Here, $N$ is the number density of color centers, $\Delta_{\omega}^\gamma$ is a pump detuning, $\mu_{ij}$ [$\gamma_{ij}$] denotes a dipole moment [a dephasing rate] of optical transition between energy levels, $|i\rangle$ and $|j\rangle$, and $\Omega_{\alpha} = \sqrt{\omega_{\alpha}^2 - (\gamma_{12} + \gamma_{13}/2)^2}$ and $\gamma_{z} = (\gamma_{12} + \gamma_{13}/2)$ are an effective Rabi frequency and an effective dephasing rate, respectively [33]. We then obtain a biphoton state function

$$|\Psi_{\text{em}}(\tau_1, \tau_2)\rangle = \psi(\tau) \exp[-i(\omega_{\alpha} \tau_1 + \omega_{\beta} \tau_2)],$$

where

$$\psi(\tau) = (L/2\pi) \int d\omega \kappa(\omega_{\alpha}, \omega_{\beta}) \Phi(\omega_{\alpha}) e^{i\omega_{\alpha} \tau_1}$$

is a two-photon wavefunction, $\Phi(\omega_{\alpha}) = \text{sgn}(\Delta k L / 2) e^{i(\omega_{\alpha} + k \cdot \vec{r})}$ is a longitudinal detuning function, $\omega_{\alpha}$ is a central frequency value, and $\tau = \tau_2 - \tau_1$ denotes a time delay. By considering the atom-like solid-state model as illustrated in Fig. 1(b), we evaluate a biphoton emission rate $R$ (sec$^{-1}$) as

$$R = \int \left| \psi(\tau) \right|^2 = \frac{L}{2\pi} \int \mid \omega_{\alpha} \mid \kappa(\omega_{\alpha}) \Phi(\omega_{\alpha})^2.$$  

Biphoton coincident counts are proportional to $R$ in Eq. (3). To experimentally demonstrate a heralded single photon pair generation, one needs to confirm nonclassical properties like the violation of the Cauchy–Schwarz inequality [17]. Our proposed scheme here focuses on the level of biphoton generation and the criteria of conventional atomic physics theory applicable to the case of solid-state parameters.

Applying the aforementioned theoretical formalism, we compare parameters of cold Rb atoms as a reference with our proposed parameters for SiV−, as shown in Table 1. We choose parameters of SiV− similar to the cold atomic ensemble [15,33] except for the linewidth $\gamma_{12}$ and ground state dephasing $\gamma_{13}$. We assume an ideal situation for SiV− ensemble as a collection of noninteracting single defects to estimate $\gamma_0$. The ground state dephasing $\gamma_{12}$ is determined by the spin coherence time $T^*_{\gamma} \approx 40$ ns in recent coherence population transfer experiments [34,35]. $\lambda_{\alpha}$ [$\lambda_{\beta}$] represents a wavelength of an anti-Stokes photon [a Stokes photon]. The ground state splitting is $\Delta_{hf} = 46$ GHz, as described in the beginning. We let $L = 1$ mm for SiV− as a proposed sample size, which can be extended by an appropriate sample fabrication process, while $L = 1$−2 cm for cold Rb atoms. $N = 10^{12} \sim 10^{18}$ cm$^{-3}$ is estimated according to 1 ppm, a relatively low defect density but an order of magnitude higher than cold atoms ($N = 10^{10} \sim 10^{17}$ cm$^{-3}$). For the maximum number density case, $N = 10^{18}$ cm$^{-3}$, the average distance between two emitters is about 10 nm (about 200 Bohr radius) under other effects such as interactions between spins or dipoles are negligible. Consequently, optical depths (OD) are given by $N\sigma_{\alpha} L$, where $\sigma_{\alpha} = (10^{-14} \sim 10^{-13}$ cm$^2$) denotes the absorption cross section. An absorption cross section or a dipole moment ($\mu_{12} \times 10^{-29}$) of SiV− can be estimated from absorption or photoluminescence (PL) measurement. Hence, we set $\sigma_{13}$ and $\mu_{13}$ as the same order of cold atomic cases.

### Table 1. Comparison of Parameters for Cold Atoms and SiV−

<table>
<thead>
<tr>
<th></th>
<th>Cold Rb Atoms [15,33]</th>
<th>SiV− [28,30]</th>
</tr>
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<tbody>
<tr>
<td>OD</td>
<td>11</td>
<td>53</td>
</tr>
<tr>
<td>$\lambda_{\alpha}$</td>
<td>794.76 nm</td>
<td>736.9118 nm</td>
</tr>
<tr>
<td>$\lambda_{\beta}$</td>
<td>780.24 nm</td>
<td>736.5293 nm</td>
</tr>
<tr>
<td>$\Delta_{hf}$</td>
<td>3.0 GHz</td>
<td>46 GHz</td>
</tr>
<tr>
<td>$\Gamma_0$</td>
<td>6 MHz</td>
<td>160 MHz</td>
</tr>
<tr>
<td>$\gamma_{12}$</td>
<td>3 MHz</td>
<td>80 MHz</td>
</tr>
<tr>
<td>$\gamma_{13}$</td>
<td>0.675 GHz</td>
<td>0.057 GHz</td>
</tr>
<tr>
<td>$\Omega_{\alpha}$</td>
<td>23.40 $\gamma_{13}$</td>
<td>4.09 $\gamma_{13}$</td>
</tr>
<tr>
<td>$\gamma_{z}$</td>
<td>0.8 $\gamma_{13}$</td>
<td>0.51 $\gamma_{13}$</td>
</tr>
<tr>
<td>$\text{BW}(\text{EIT})$</td>
<td>247.7 MHz</td>
<td>3.64 MHz</td>
</tr>
<tr>
<td>$\text{BW}(\text{PM})$</td>
<td>412.9 MHz</td>
<td>2.76 MHz</td>
</tr>
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As discussed earlier and shown in Fig. 1, we set a natural linewidth (full-width at half-maximum, FWHM) $\Gamma_0$ and a dephasing rate $\gamma_{13}$ between energy levels 1 and 3. For driving fields, we set the power of a coupling ($P_c = 180$ mW) and a pump laser ($P_p = 3$ mW), and consequently the Rabi frequencies ($\Omega_c = 4.78\gamma_{13}$ and $\Omega_p = 1.45\gamma_{13}$) and the effective dephasing rate ($\gamma_e$). A far off-detuned pump beam $\Delta_p = 48.67\gamma_{13}$ is a similar condition in a cold atomic case. Finally, to characterize biphoton waveforms in Fig. 2, we list bandwidths (BW) of EIT and a phase matching (PM) that agree with the values in Fig. 3(a).

To begin, we reproduce biphoton waveforms of a reference system (Figs. 2(a), OD = 1.1, and 2(c), OD = 53, of Ref. [33]) by using parameters of cold Rb atoms [15,33] in Table 1. We then evaluate $|\psi(\tau)|^2$ (sec$^{-2}$) for our proposed SiV$^+$ parameters as in the first row of Fig. 2 for four different OD cases: (a) OD = 1.1, (b) 11, (c) 53, and (d–e) 107. Figures 2(a)–2(b) [(c–e)] show a low OD Rabi oscillatory regime [a high OD group delay regime demonstrating a square-like wave packet]. Although the timescale is only tens of nanoseconds for the SiV$^+$ case, the waveforms are similar to cold atomic cases, especially for high OD [33]. We further investigate the behavior of wavepackets from SiV$^+$ by looking at the spectral properties of each part. We evaluate the biphoton spectra $|\kappa(\omega_{\omega_p})\Phi(\omega_p)|^2$ in the second row, and anti-Stokes EIT spectra in the third. Figure 2(a) shows a 2.7 ns period of an oscillatory feature on the biphoton wavepacket corresponding to the distinct two spectral peaks separated by $\Omega_e = 4.68\gamma_{13}$ ~ 372 MHz that appears in the second row. Here, the FWHM
linewidth of each spectral sideband agrees with the effective dephasing rate \( \gamma_d = 0.52\gamma_{13} \approx 40 \text{ MHz} \). Both the spectrally wide EIT window and partially absorptive spectrum indicate that the characteristics of the low OD is 1.1.

As we increase the OD, however, two peaks in the biphoton spectrum coalesce to form a single peak at the resonance, especially in the group delay regime \([\text{OD} > 50, \text{Figs. 2(c)–2(e)}]\). Consequently, the biphoton spectrum becomes narrower, and the EIT transmission peak decreases due to a non-negligible dephasing \( \gamma_{12} \). Figure 2(e) shows the case of zero ground state dephasing \( \gamma_{12} = 0 \) to demonstrate an ideal EIT transmission compared to the \( \gamma_{12} = 0.05\gamma_{13} \) case in Fig. 2(d). To investigate \( \gamma_{12} = 0 \) and \( \Omega_d \) dependence, we plot EIT [PM] bandwidths in Fig. 3 that are inversely proportional to \( \sqrt{\text{OD}} \) [OD] [33]. The PM bandwidth becomes narrower than the EIT for OD > 30 so that, for Fig. 3(a), the EIT [PM] bandwidth becomes 87.7 MHz [46.7 MHz] as shown in Fig. 2(d) and Table 1, which is less than 100 MHz. Note that biphoton spectrum BW agrees well with EIT BW for high OD cases.

To further decrease the EIT bandwidth and eventually achieve below subnatural atomic linewidth in a solid-state sample, we consider a weak coupling \( \Omega_d = 1.07\gamma_{13} \) and negligible \( \gamma_{12} = 0 \) as in Fig. 3(b). Figure 4 shows a group delay time \( \tau_d = L/V_s = L(dk_d/d\omega) \) at OD = 107 for such a case, which shows a several hundreds nanosecond coherence time of wavepackets, a characteristic of the subnatural linewidth. A realization of nearly zero ground state dephasing \( \gamma_{12} = 0 \) would be the key to achieve a narrowband below atomic natural linewidth, 6 MHz.

Finally, in addition to our proposed SiV− in the diamond system, we apply our suggested criteria to other solid-state candidates, such as nitrogen-vacancy (NV−) defect centers [36,37], germanium-vacancy (GeV−) defect centers [38,39], and tin-vacancy (SnV−) in diamond [40] as shown in Table 2. SiV−, GeV−, and SnV− (group IV in the periodic table) with \( D_{3h} \) structural symmetry exhibit their sharp spectral properties and the four distinct energy levels compared to the NV defect diamond; consequently, they are the most appropriate candidates. The drawback, however, might be blinking of the fluorescence, which can be improved by the sample growth technique [39]. Although we did not include them in Table 2, quantum dots or hybrid two-dimensional materials can be categorized with various energy schemes of the EIT-SFWM as investigated in the cold atomic system.

In conclusion, we propose the observability of EIT-SFWM-based biphoton generation in an atom-like solid-state system, SiV−, towards long-distance quantum communication by entangled photons. We find the matching criteria of solid-state biphoton generation from SiV− where we can directly use existing biphoton theory of the cold atomic ensemble. Such criteria include crystal structure affecting the broadening and polarization, crystal axis associated with the polarization condition in SFWM, and the selection rule for an optical transition. In detail, two excited states and two ground states associated with strong optical transitions in SiV− form a double \( \Lambda \) system as a basis of Stokes and anti-Stokes photon pair generation. Spin conserving relaxation from excited states to ground states determines the selection rule for each dipole transition. Under the zero magnetic field, intrinsic light polarizations of the solid-state samples are characterized by the crystal axis, which matches our design of four-wave mixing: H-polarized light for two driving fields and V-polarized light for Stokes and anti-Stokes photons. The main challenge (or difference) of applying the atomic model to solid-state systems is to estimate absolute values of absorption coefficients \( \alpha_0 \) and theoretical dipole moments \( \mu_j \). Therefore, we consider empirical weights based on existing spectral studies of SiV−. Several merits to using solid-state samples include high OD if we overcome the inhomogeneous broadening issues and reduce the ground state dephasing \( \gamma_{13} \) further. Enhanced light coupling efficiency of the nanowire array [29] would be useful in our proposed narrowband biphoton generator. Our proposed EIT-SFWM-based diamond defects photon source is ideal when it is combined with a spin-based memory in a way that photondriven ground state spin flips, so that the spin state controls biphoton generation for a read-write process of quantum information. Our approach might pave a way toward a 1 μs long coherence time for practical usage of solid-state based quantum communication.

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**Table 2. Parameters for Quantum Emitter Candidates: Nitrogen-Vacancy (NV−), Silicon-Vacancy (SiV−), and Germanium-Vacancy (GeV−) Center of Diamond Defects**

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<tr>
<td>( \lambda_d )</td>
<td>637.199 nm</td>
<td>736.9118 nm</td>
<td>602.54 nm</td>
</tr>
<tr>
<td>( \lambda_r )</td>
<td>637.189 nm</td>
<td>736.5293 nm</td>
<td>601.54 nm</td>
</tr>
<tr>
<td>( \Delta_f )</td>
<td>2.88 GHz</td>
<td>46 GHz</td>
<td>152 GHz</td>
</tr>
<tr>
<td>( \Gamma_0 )</td>
<td>16 MHz</td>
<td>160 MHz</td>
<td>52 MHz</td>
</tr>
<tr>
<td>( \gamma_{13} )</td>
<td>8 MHz</td>
<td>80 MHz</td>
<td>26 MHz</td>
</tr>
<tr>
<td>( \gamma_{12} )</td>
<td>1.0–2.4 kHz</td>
<td>4 MHz</td>
<td>9 MHz</td>
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**Fig. 4.** Group delay \( \tau_d \) as a function of \( \Omega_d \) for zero dephasing \( \gamma_{12} = 0 \) and OD = 107. Insets show that \( \tau_d \) agrees with coherence timescale in the high OD group delay regime.

**References**
