

Quantum Orbital-State Control of a Neutral Nitrogen-Vacancy Center at Millikelvin Temperatures

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(Received 27 November 2024; revised 22 January 2025; accepted 21 May 2025; published 2 July 2025)

A neutral nitrogen-vacancy center (NV⁰) is promising for realizing strong coupling with a single microwave photon due to its large electric field sensitivity, although it is susceptible to environmental phonon noise at 5 K. Decreasing the temperature to 15 mK results in a tenfold increase in orbital relaxation time compared to that at 5 K. Dynamical decoupling pulses significantly increase the orbital coherence time to more than 1.6 μ s, representing a 30-fold improvement compared to that without decoupling pulses. Based on these results, a single NV⁰ can reach the strong coupling regime when coupled with a high-impedance microwave resonator, thus opening up the possibility of microwave quantum electrodynamics using a single optically active defect center in diamond.

DOI: 10.1103/m1qx-xlpd

Diamond color centers have been intensively studied due to their potential applications as quantum sensors [1], single-photon sources [2–5], quantum processors [6–9], and quantum repeaters [10–14]. Among these color centers, the negatively charged nitrogen-vacancy center (NV[−]) is the most popular and well studied due to its high quantum efficiency and the ability to operate at room temperature [7,15–19]. Recently, however, group IV color centers characterized by higher zero-phonon line emission efficiency and better spectral stability in a nanostructure have gained increasing attention in applications for quantum communication [19–28]. Research into novel diamond color centers for specific applications is still actively underway.

A neutral nitrogen-vacancy center, NV⁰, has an orbital degree of freedom in its ground state, resulting in a five orders of magnitude larger electric field [29] and strain [30] susceptibility compared to the ground state of NV[−] [31,32]. Since the large susceptibility to environmental phonons

results in short orbital relaxation time (several hundred nanoseconds at 5 K) [33], NV⁰ has not attracted as much interest as NV[−]. However, its large electric field response and ground-state splitting around 10 GHz [29,33] are advantageous when considering the orbital state control using microwave electric fields and realization of microwave cavity quantum electrodynamics using a single color center in diamond. While strong coupling to an optical photon has been achieved in a silicon vacancy center [34], and strong coupling to a flux qubit has been realized with an ensemble of NV[−] centers [35], strong coupling to a microwave field using a single color center remains a challenge. It is noteworthy that the coupling to microwave resonators is actively investigated in other solid-state quantum systems [36–43].

In this Letter, we perform quantum control of the orbital state of NV⁰ and estimate the orbital relaxation and coherence times at millikelvin temperatures. We observe an exponential increase in the orbital relaxation time due to a decrease in environmental phonons. Additionally, the orbital coherence is extended through the application of dynamical decoupling techniques. Based on the obtained relaxation and coherence times, strong coupling between NV⁰ and a high-impedance microwave resonator [44,45] is achievable, suggesting the possibility of quantum electrodynamics experiments using an optically active diamond color center, such as controlling and reading out NV⁰ using a single microwave photon as well as optical photons.

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In the following, all measurements are performed in a dilution refrigerator with a homebuilt confocal microscopy system. The temperature at the mixing chamber plate, T_{MxC} , is maintained at 50 mK unless specified. A diamond sample is mounted onto a stack of three-axis piezo positioners and a three-axis piezo scanner. A 10- μm -diameter solid-immersion lens is fabricated on the surface of the diamond to enhance the collection efficiency. Driving electric fields are applied using Au/Ti electrodes formed on the sample (see Supplemental Material [46]).

First, we briefly introduce the energy level structure of NV^0 and show experiments characterizing the energy level of NV^0 . In the ground state, NV^0 has four energy levels with orbital ($|\pm\rangle_o$) and spin ($|\uparrow\rangle_s, |\downarrow\rangle_s$) degrees of freedom [Fig. 1(a)], which are the eigenstates of the Hamiltonian [29,33] in the weak strain limit ($\lambda \gg \epsilon_\perp$),

$$\hat{H}^{(\pm)}/\hbar = g_s \mu_B B_z \hat{S}_z + g_l \mu_B B_z \hat{L}_z + 2\lambda \hat{L}_z \hat{S}_z + \epsilon_\perp (\hat{L}_+ + \hat{L}_-), \quad (1)$$

where g_s is the spin g factor, μ_B is the Bohr magneton, g_l is the orbital g factor, B_z is the magnetic flux density, $\hat{S}_z = \hat{\sigma}_z/2$ is the half-spin operator, $\hat{L}_z = \hat{\sigma}_z$ is the orbital operator for $|\pm\rangle_o$ basis, $|\pm\rangle_o = \mp(|e_x\rangle_o \pm i|e_y\rangle_o)$, and $|e_x\rangle_o$ and $|e_y\rangle_o$ are the orbital states of the nitrogen vacancy [33,47]. Considering the static strain term in the Hamiltonian, $\epsilon_\perp (\hat{L}_+ + \hat{L}_-)$, the spin-orbit coupling, λ , and the strain, $\epsilon_\perp$, cause the ground-state splitting to the spin-orbital state to be $\Delta_{\text{gs}} = 2\sqrt{\lambda^2 + \epsilon_\perp^2}$ [29,33,50] in its ground state. Here, $\hat{L}_\pm$ is the orbital raising and lowering operators, which hybridize $|\pm\rangle_o$ (see Supplemental Material [46] for detailed explanation of the Hamiltonian). Under an ambient magnetic field of 0.01 mT (in our setup), the spin and orbital states are nearly degenerate. We confirm that this environmental magnetic field has a negligible impact on our experiment (see Supplemental Material [46] for detailed discussions on the effects of stray static magnetic fields and of microwave magnetic fields induced by the driving microwaves). Thus, in the following experiments, we ignore the spin degree of freedom and treat it as an effective three-level system. We denote the lower energy level as $|0\rangle$, the upper energy level as $|1\rangle$, and the optically excited level as $|2\rangle$.

Regarding the optical properties of NV^0 , the optical excitation occurs under illumination with a 575-nm laser. After irradiating a 637-nm laser resonant with the zero-phonon line of NV^- for charge initialization, the frequency of the 575-nm laser is swept to obtain a photoluminescence excitation (PLE) spectrum [Fig. 1(b)]. The two observed peaks are identified as $|0\rangle \rightarrow |2\rangle$ and $|1\rangle \rightarrow |2\rangle$ transitions, respectively. The separation between two peaks corresponds to $\Delta_{\text{gs}}/2\pi$, which is $13,089 \pm 9$ MHz [Fig. 1(c)]. Assuming that $\lambda/2\pi = 4.9$ GHz [33], $\epsilon_\perp/2\pi = 4.3$ GHz for the measured NV^0 . The minimum PLE linewidth

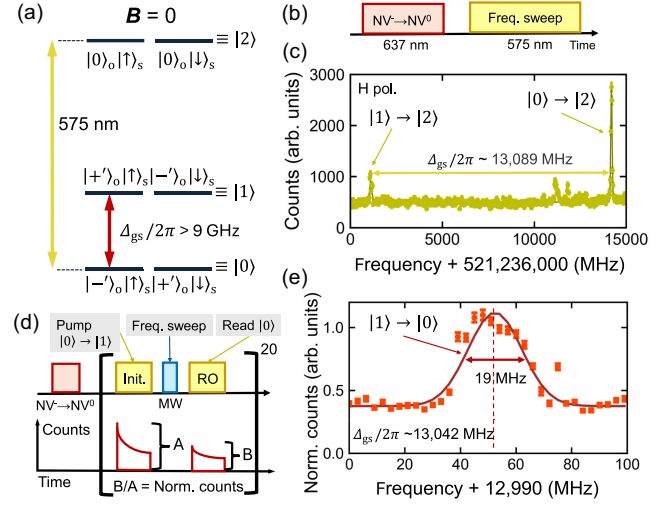


FIG. 1. Energy level structure of NV^0 and its characterization. (a) Schematic of the energy levels of NV^0 with some static strain under a zero magnetic field. $|\pm\rangle_o$ and $|\uparrow\rangle_s$ are the eigenstate of the orbital (o) and spin (s) operators. The prime symbol ($|\pm'\rangle$) indicates the orbital eigenstates are hybridized under the static strain [29]. $|0\rangle_o$ represents that the orbital angular momentum is zero. (b) Measurement sequence for PLE spectrum of NV^0 . The 637-nm laser that is resonant with the zero-phonon line of NV^- is used to initialize the charge state to NV^0 , while the 575-nm laser is used to observe the optical transition. (c) PLE spectrum of NV^0 . The 575-nm laser is horizontally polarized to maximize counts from $|0\rangle$. (d) Measurement sequence for ODER. After the charge initialization, the driving microwave pulse (30 ns) to invert the population is applied between initialization (Init.) and readout (RO) optical pulses. The initialization pulse pumps the orbital state from $|0\rangle$ to $|1\rangle$. Changes in population induced by the driving pulse are estimated from the ratio of the maximum counts (B/A) obtained from the first (A) and second (B) pulses. In the following analysis, the dark counts are subtracted from the PL counts. The PL counts are then smoothed by the five-point moving average. The number, 20, above the parenthesis corresponds to the repetition times before applying another NV^0 initialization pulse (see Supplemental Material [46] for details). The total number of the repetition is in the order of 10^7 . (e) The ODER spectrum corresponding to the $|1\rangle \rightarrow |0\rangle$ transition, which is fitted by a Gaussian curve. The error bars correspond to the standard deviation considering the photon shot noise.

(FWHM) is 44.8 MHz (see Supplemental Material [46]), which is approximately six times larger than the Fourier-transform-limited linewidth (of 7.2 MHz [33]).

We also perform an optically detected electric resonance (ODER) experiment to confirm the ground-state resonance frequency using driving microwave electric fields as shown in Figs. 1(d) and 1(e). The center frequency is 13 042 MHz ($= \Delta_{\text{gs}}/2\pi$), and the FWHM is 19 ± 0.4 MHz from the Gaussian fit. Δ_{gs} is in good agreement with the value obtained from the PLE measurement. The slight deviation in the estimation of Δ_{gs} between PLE and ODER is attributed to the limited frequency resolution in the PLE

measurement and the frequency drift during the measurement. The following measurement reveals that the Rabi frequency for this measurement is 14 MHz, limiting the ODER linewidth by the power broadening. However, due to spectral hopping, it is difficult to further reduce the ODER linewidth by decreasing the driving microwave power.

At millikelvin temperatures, the orbital relaxation time, T_1^{orb} , of NV^0 is expected to increase compared to that at 5 K since phononic excitations around 10 GHz in the environment are reduced by several orders of magnitude. T_1^{orb} is estimated using the pump-probe technique similar to the ODER measurement. As expected, T_1^{orb} increases from 150 ± 60 ns at $T_{\text{MXC}} = 8.0$ K to 4.7 ± 0.4 μs at $T_{\text{MXC}} = 10$ –15 mK [Fig. 2(a)]. The exponential increase in T_1^{orb} is in reasonably good agreement with a theoretical curve assuming a relaxation via a single-phonon process, $1/T_1^{\text{orb}} = A\Delta_{\text{gs}}^3(2n_{\text{BE}}(\Delta_{\text{gs}}, T) + 1)$ [51], where A is a constant and n_{BE} is the average population in the Bose-Einstein distribution. The reasonable agreement with the fitting curve indicates that T_{MXC} is close to the sample temperature. The exponential increase in T_1^{orb} is explained by the existence of the ground-state splitting (energy gap). When the

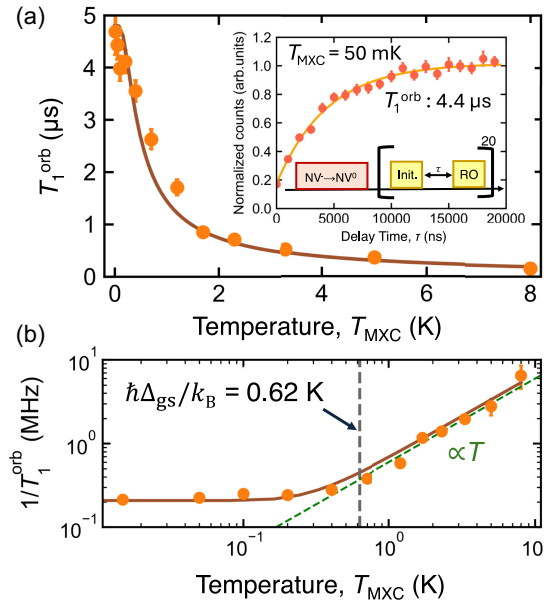


FIG. 2. Temperature dependence of the orbital relaxation time and rate. (a) Temperature dependence of the orbital relaxation time. The fitting function for T_1^{orb} is shown in the main text. The inset shows how T_1^{orb} is extracted from the measurement of the population recovery after the pumping pulse from $|0\rangle$ to $|1\rangle$ at $T_{\text{MXC}} = 50$ mK, where T_{MXC} is the temperature of the mixing chamber plate. The error bars correspond to the standard error in the nonlinear fitting. The measurement sequence is also shown in the inset, representing that the time delay between the initialization and readout pulses is swept. (b) Temperature dependence of the orbital relaxation rate. The green dashed line is proportional to temperature.

ground-state splitting exceeds the thermal energy, $k_{\text{B}}T$, with decreasing temperature, thermal excitation is suppressed exponentially, resulting in the increase in T_1^{orb} . At $T_{\text{MXC}} > 0.62$ K, $1/T_1^{\text{orb}}$ increases linearly with temperature as shown in Fig. 2(b), indicating that the single-phonon process is dominant. The linear temperature dependence at higher temperatures is consistent with the report in Ref. [33].

After characterizing the orbital relaxation time, we proceed to perform coherent control of the orbital states and estimate the orbital coherence time. Following the orbital initialization from $|0\rangle$ to $|1\rangle$ using a 575-nm pulse, a microwave pulse resonant with Δ_{gs} is applied with changing the width. The time delay of the readout pulse is also adjusted as the pulse width increases. Rabi oscillation between $|0\rangle$ and $|1\rangle$ with a decaying time constant (T_2^{Rabi}) of 46 ± 2 ns is observed as shown in Fig. 3(a). Maximum Rabi frequency is 27 MHz with an input peak power of 3.9 mW (average power: 41 μW outside the fridge, 10 μW at the sample for the 180-ns pulse), which is measured at outside the dilution refrigerator (see Supplemental Material

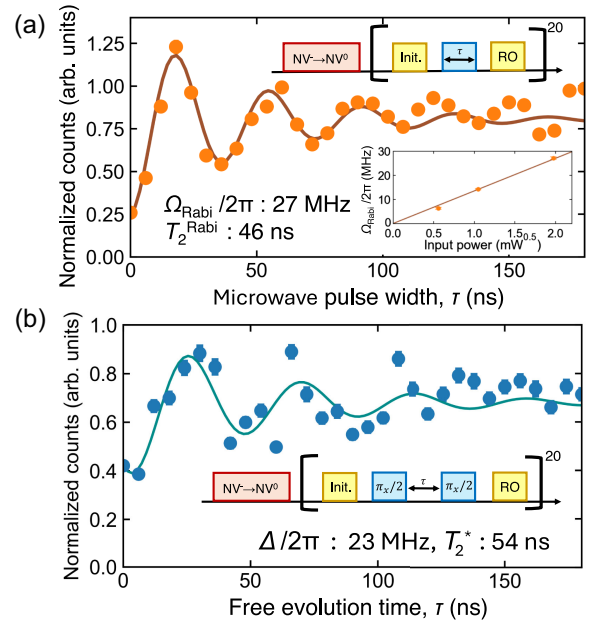


FIG. 3. The Rabi oscillation and the Ramsey interference with their measurement sequences. (a) The Rabi oscillation between the orbital states, $|0\rangle$ and $|1\rangle$. The driving power outside the dilution refrigerator is 3.9 mW. The data are fitted with a function, $A \cos(\Omega_{\text{Rabi}}t) \exp(-t/T_2^{\text{Rabi}}) + B$, where A is the amplitude, B is the center of the oscillation, Ω_{Rabi} is the Rabi frequency. The inset shows the Rabi frequency as a function of the square root of the driving power. The error bars correspond to the standard deviation considering the photon shot noise. (b) Ramsey interference when the driving frequency is set to be 13 020 MHz. The fitting function is $C \cos(\Delta t + \phi) \exp(-t/T_2^*) + D$, where C is the amplitude, Δ is the detuning from the resonant frequency, ϕ is the phase offset, and D is the center of the oscillation. The error bars correspond to the standard deviation considering the photon shot noise.

[46] for the microwave induced heating). The Rabi frequency is proportional to the square root of the input power as shown in the inset of Fig. 3(a), indicating that the detuning is negligibly small. Additionally, the initialization fidelity is estimated to be $85 \pm 3\%$ (see Supplemental Material [46]).

The orbital coherence time, T_2^* , is estimated to be 54 ± 5 ns from the Ramsey interference measurement [Fig. 3(b)] with a certain detuning from Δ_{gs} . The value of T_2^* is close to that of T_2^{Rabi} , indicating that similar noise processes can dominate T_2^* and T_2^{Rabi} . According to Ref. [29], T_2^* at 5 K is 31 ± 3.6 ns. In contrast to the orbital relaxation time, the orbital coherence time does not increase with decreasing temperature, indicating it is not limited by the thermal phonon in the environment.

We also apply the dynamical decoupling techniques to extend the orbital coherence time of NV^0 . By applying bit-flipping pulses [Fig. 4(a)], the dynamically decoupled coherence time, $T_2^{\text{echo/CPMG}}$, increases to 1.6 ± 0.3 μs for Hahn-echo and up to 2.0 ± 0.5 μs for the Carr-Purcell-Meiboom-Gill (CPMG) sequence [Figs. 4(b) and 4(c)]. More than a 30-fold increase in the orbital coherence time is achieved by applying these decoupling techniques. Since the dynamical decoupling sequences filter out low-frequency phase noise throughout the entire measurement [52], this improvement indicates that the dominant noise source is non-Markovian, such as spectral diffusion and fluctuations, which occur on a relatively slow timescale compared to $T_2^{\text{echo/CPMG}}$.

The enhancement in $T_2^{\text{echo/CPMG}}$ is modest with increasing the number of decoupling pulses [Fig. 4(c)], which is attributed to the decrease in T_1^{orb} down to 1.3–1.8 μs due to the local heating from the driving microwave (see Supplemental Material [46]). Especially for CPMG 8, $T_2^{\text{echo/CPMG}}$ approaches $2T_1^{\text{orb}}$, which ultimately limits the coherence time. It should be noted that since assuming the exponential decay for $T_2^{\text{echo/CPMG}}$ is not always applicable, we discuss dependence on the fitting function and the dephasing time, $T_\phi^{\text{echo/CPMG}}$, in Supplemental Material [46].

Based on the relaxation and coherence times obtained from the above measurements, we explore the possibility of realizing cavity quantum electrodynamics using NV^0 and a microwave photon. The single microwave photon voltage generated by the state-of-the-art microwave resonator with 4-k Ω impedance [44,45] is $\omega_{\text{mw}}\sqrt{\hbar Z/2} \sim 37$ μV , where $\omega_{\text{mw}}/2\pi = 13$ GHz is the angular frequency of the microwave resonator, Z is the characteristic impedance of the microwave resonator. Considering that NV^0 is placed between the gap of a ring microwave resonator, the electric field between the gap is $37 \times 2/d$ $\mu\text{V}/\text{m}$, where d is the distance between the gap (see Supplemental Material [46] for a schematic). The factor of 2 is introduced because one end of the ring resonator has +37 μV and the other end has

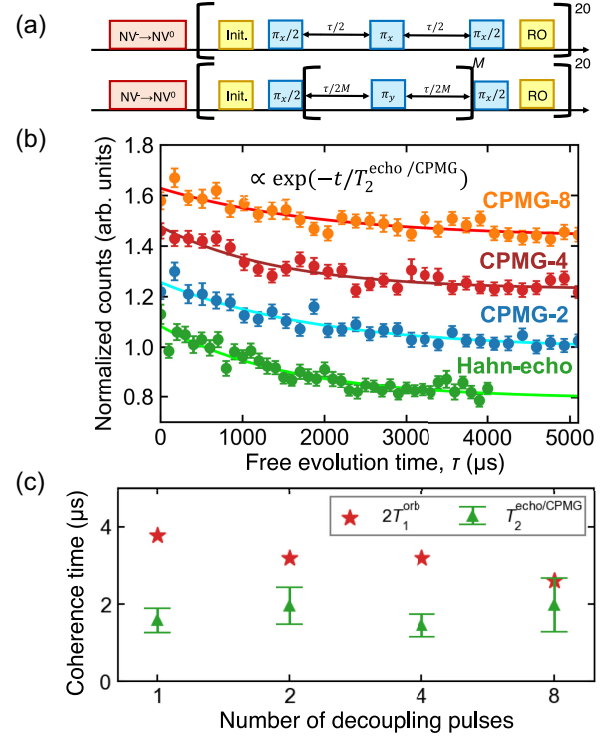


FIG. 4. Coherence extension by dynamical decoupling. (a) The pulse sequences for Hahn echo (top) and Carr-Purcell-Meiboom-Gill (CPMG) M (bottom). The lowercase notation $\pi_{x(y)}$ indicates that the phase of bit-flipping pulse is in(quadrate) phase. (b) Coherence decay under the application of dynamical decoupling pulses [Hahn echo, CPMG M ($M = 2, 4, 8$)], with offsets applied to each curve. The decaying curves are fitted with the function, $E \exp(-t/T_2^{\text{echo/CPMG}}) + F$, where E and F are some constants. The error bars correspond to the standard deviation considering the photon shot noise. (c) The number of the decoupling pulses versus the extended coherence time. The error bars correspond to the standard error in the nonlinear fitting. The upper limit of the coherence, $2T_1^{\text{orb}}$, is also shown (see Supplemental Material [46] for the pulse-number dependence of $2T_1^{\text{orb}}$).

−37 μV . When we set d to be 1 μm , the resulting electric field is 0.74 V/cm. Since the electric field sensitivity of NV^0 is 1 MHz/V cm^{−1} [29], the coupling strength, $g_{\text{mw-NV}^0}/2\pi$, between the microwave electric field and NV^0 is 740 kHz. Assuming the relaxation rate of NV^0 , $\gamma_1^{\text{NV}^0}/2\pi$, to be 34 kHz ($= 1/2\pi T_1^{\text{orb}}$), and that of the microwave resonator, $\kappa_{\text{mw}}/2\pi$, to be 100 kHz ($Q_{\text{mw}} = 13 \text{ GHz}/100 \text{ kHz} = 1.3 \times 10^5$), the strong coupling regime, in which $g_{\text{mw-NV}^0} > \gamma_1^{\text{NV}^0}, \kappa_{\text{mw}}$ and $C = 4g_{\text{mw-NV}^0}^2/\gamma_1^{\text{NV}^0}\kappa_{\text{mw}} = 664$, is achievable, where C is the cooperativity. Even stronger coupling can be achieved when NV^0 is placed in a phononic crystal cavity [54], although the integration into a nanostructure like a phononic crystal presents challenges, as it can induce significant spectral diffusion for NV centers.

In conclusion, we have demonstrated quantum control of NV^0 in a dilution refrigerator and have measured the orbital

relaxation and coherence times. The orbital relaxation time has shown an approximately tenfold increase compared to that at 5 K. The orbital coherence time has not exhibited an improvement just by decreasing the temperature. However, we have extended the coherence time to more than 1.6 μ s using the dynamical decoupling. This is a 30-fold increase compared to the coherence time without the dynamical decoupling. Based on these results, the strong coupling regime between a single NV⁰ and a high-impedance microwave resonator or a phononic crystal cavity is within reach. Compared to group IV color centers, the relatively small Δ_{gs} (~ 10 GHz) and high electric field sensitivity of NV⁰ are advantages realizing strong coupling to other fields. Although the stabilization of the spectrum remains a challenge, this Letter opens the possibility of using the optically active defect center, NV⁰, as a quantum interface between different degrees of freedom: microwave photon, microwave phonon, and optical photon.

Acknowledgments—H. Kosaka acknowledges the funding support from Japan Science and Technology Agency (JST) Moonshot R&D Grant No. JPMJMS2062 and JST CREST Grant No. JPMJCR1773. H. Kosaka also acknowledges the Ministry of Internal Affairs and Communications (MIC) for funding, research and development for construction of global quantum cryptography network (JPMI00316), and the Japan Society for the Promotion of Science (JSPS) Grants-in-Aid for Scientific Research (20H05661, 20K20441).

H. Kurokawa designed and performed the experiments. S. Nakazato also performed experiments in a different sample. T. Makino and H. Kato fabricated the electrical circuit. S. Onoda contributed to the creation of NV centers by the electron beam irradiation and annealing. Y. Sekiguchi contributed the fabrication of the solid-immersion-lens structure. H. Kosaka supervised the project.

The authors declare no competing interests.

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