

## Hot-Phonon-Induced Distortion of Diamond Defects on Ultrafast Timescales

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(Received 1 May 2025; revised 29 August 2025; accepted 20 October 2025; published 18 November 2025)

We investigated ultrafast defect-lattice dynamics in diamond using the  $N_s:H-C^0$  defect, an analog of bond-centered hydrogen in semiconductors. Combining synthesis, ultrafast vibrational spectroscopy, and *ab initio* calculations, we show that excitation of the defect's stretch mode leads to the generation of localized phonons and the formation of a hot ground state, where the interatomic potential is transiently modified. Our results reveal unexpected nonequilibrium phonon effects despite diamond's exceptionally high thermal conductivity, with implications for quantum defect engineering.

DOI: 10.1103/mvdf-bdrx

Diamond hosts important functional defects such as the nitrogen-vacancy (NV) and silicon-vacancy (SiV) centers, which have promising applications in quantum information and nanoscale magnetometry due to their long spin coherence times and ability to be optically manipulated [1–3]. High-precision engineering and control of such defects require a deep understanding of the defect-lattice interactions that govern energy relaxation, particularly the rate at which thermal energy dissipates following excitation. Diamond's rigid C-C covalent network yields an exceptional thermal conductivity ( $\kappa = 2200 \text{ W m}^{-1} \text{ K}^{-1}$  at room temperature), suggesting efficient heat removal from atomic-scale defects following vibrational excitation. In the classical, macroscopic picture, diamond's high thermal diffusivity  $\alpha = \kappa/C_V$  implies rapid smoothing of temperature gradients via the heat equation  $\partial T/\partial t = \alpha \nabla^2 T$ . Microscopically, however, the discrete local vibrational modes (LVMs) of a defect must couple to lattice phonons to relax.

We consider a three-stage process to transfer thermal energy from an excited molecular defect to its host solid, as pictured in Fig. 1(a). In the first stage, a defect is vibrationally excited by a photon or phonon (left panel). In stage

2, the LVM relaxes by phonon emission, distorting the local atoms from their equilibrium positions (center panel)—a process akin to polaron formation in semiconductors. In

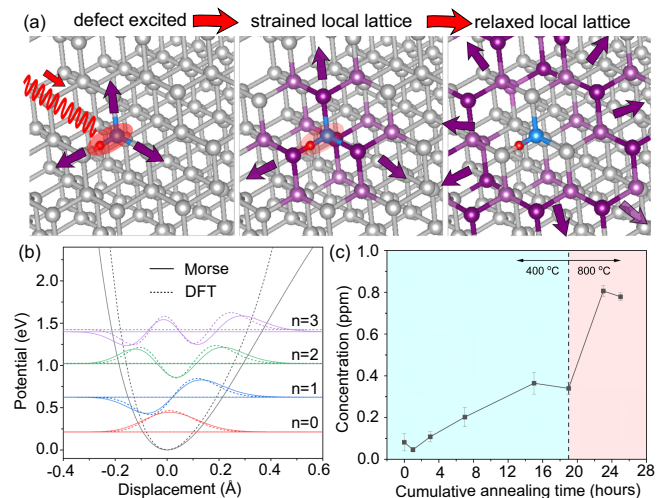


FIG. 1. (a) Excitation and relaxation of the  $N_s:H-C^0$  defect in a diamond crystal. Gray, blue, and red spheres represent carbon, nitrogen, and hydrogen atoms, respectively. Purple arrows show the transfer of energy away from the defect. (b) Calculated vibrational mode energies and wave functions for the  $N_s:H-C^0$  C-H bond stretch mode, using the Morse potential (solid line) and first-principles DFT calculations (dashed lines), for the  $n = 0$  ground state (red) and the  $n = 1$  (blue),  $n = 2$  (green), and  $n = 3$  (purple) vibrational states. (c)  $N_s:H-C^0$  concentration after repeated annealing at 400 °C (blue) and 800 °C (red), derived from infrared absorption spectroscopy and theory.

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stage 3 (right panel), the defect has returned to its ground state as phonons have dispersed. This scenario raises key questions: How efficiently does the defect transfer energy to the local crystal? How fast does the crystalline lattice remove heat? And, critically, if energy transfer to the crystal outpaces the cooling rate of the crystal, do phonons created near the defect modify its properties? In solution-phase molecules, such energy transfer can heat the surrounding solvent, creating a “hot ground state” (HGS) with modified potential energy surfaces [4,5]. The lower thermal conductivity of liquids compared to solids (e.g.,  $0.6 \text{ W m}^{-1} \text{ K}^{-1}$  for water) means HGSs can persist for nanoseconds or longer. But are such effects prominent in molecular-scale defects in solid state systems, or is it valid to make the standard assumption of relaxation within a cold, static crystal?

We adopted the  $\text{N}_s\text{:H-C}^0$  defect in diamond as a model system to investigate defect-lattice interactions. The neutral substitutional nitrogen  $\text{N}_s$  impurity bonds to four neighboring carbon atoms, while the remaining unpaired electron forms a localized antibonding orbital along one N-C axis [6]. This unique N-C bond is approximately 25% longer than a normal C-C bond, with the unpaired electron mostly localized on the unique carbon. Hydrogen binds readily to this carbon, leading to the  $\text{N}_s\text{:H-C}^0$  structure [Fig. 1(a)], which exhibits a prominent LVM at  $3323 \text{ cm}^{-1}$  assigned to the C-H bond’s fundamental stretch mode [7]. This assignment is validated by (i) first-principles calculations of the interatomic potential [Fig. 1(b)] [8], (ii) isotopic substitution of nitrogen and hydrogen [9], and (iii) uniaxial stress studies [10].

The  $\text{N}_s\text{:H-C}^0$  defect is ideal for probing defect-lattice coupling: It has a simple, linear structure that minimizes internal mode coupling, such as stretch-bend interactions [11], and its similarity to a bond-centred (BC) hydrogen interstitial makes it relevant to a broad class of defects in diamond and other materials (see End Matter).

We demonstrate via ultrafast multidimensional spectroscopy that during vibrational relaxation of the  $\text{N}_s\text{:H-C}^0$  defect the production of phonons in the crystal creates a HGS despite diamond’s exceptional thermal transport properties. Density functional theory (DFT) calculations and a Morse potential model were used to simulate the C-H stretch mode potential, shown in Fig. 1(b). The transition energies for the experimentally constrained Morse potential and first-principles theory demonstrated excellent agreement: within 5% for the  $n = 2$  to  $n = 3$  transition [10], validating the use of the Morse model when deriving IR matrix elements [11,12].

$\text{N}_s\text{:H-C}^0$  centers were created in chemical vapor deposition synthetic diamonds by neutron irradiation and annealing, first at  $400^\circ \text{C}$  and then subsequently at  $800^\circ \text{C}$ , with the defect concentration tracked via the absorption of the LVM at  $3323 \text{ cm}^{-1}$  [Fig. 1(c)] and reaching 0.75 ppm

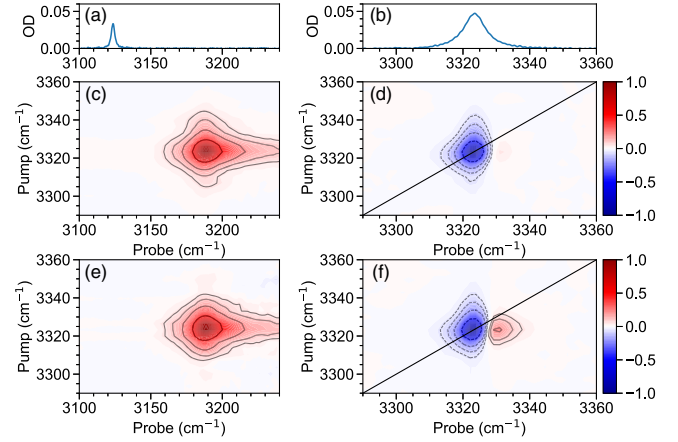


FIG. 2. Linear absorption spectra (a) and (b) between  $3100$  and  $3360 \text{ cm}^{-1}$ , containing the stretch mode of  $\text{NVH}^0$  at  $3123 \text{ cm}^{-1}$  and the stretch mode of the  $\text{N}_s\text{:H-C}^0$  defect at  $3323 \text{ cm}^{-1}$ . Spectra are shown after subtracting the background contribution of intrinsic diamond. (c) and (d) show normalized 2DIR spectrograms for the  $3188 \text{ cm}^{-1}$  ESA and  $3323 \text{ cm}^{-1}$  GSB features, respectively, measured at  $0.6 \text{ ps}$ . (e) and (f) are as (c) and (d) but at  $2.0 \text{ ps}$ . The diagonal line in (d) and (f) shows equal pump and probe energies.

[10]. We propose multiple synthesis routes for this defect based on its annealing behavior [10].

Linear absorption spectroscopy (FTIR) and multi-dimensional coherent spectroscopy (2DIR) [10,13] experiments were performed in ambient temperature on three synthetic diamonds containing the  $\text{N}_s\text{:H-C}^0$  defect (Fig. 2). The optical density (OD) from FTIR across the  $3100\text{--}3360 \text{ cm}^{-1}$  range [Figs. 2(a) and 2(b)], after subtracting the intrinsic background from three-phonon absorption, demonstrates IR resonances from the  $\text{N}_s\text{:H-C}^0$  defect stretch mode at  $3323 \text{ cm}^{-1}$  and from the  $\text{NVH}^0$  stretch mode at  $3123 \text{ cm}^{-1}$ , which coexisted in the diamond.

Pumping the  $\text{N}_s\text{:H-C}^0$  stretch and probing over the same range, we obtained 2DIR spectrograms at pump-probe delay  $t = 0.6 \text{ ps}$  [Figs. 2(c) and 2(d)] that show an on-diagonal ground state bleach (GSB) at  $3323 \text{ cm}^{-1}$  and an off-diagonal excited state absorption (ESA) at  $3188 \text{ cm}^{-1}$ . The GSB tracks the pump-induced change in ground state absorption ( $n = 0$ ), while the ESA corresponds to  $n = 1 \rightarrow 2$  and, therefore, monitors the population  $N_1$  in state  $n = 1$ . The energy difference between ESA and GSB yields an anharmonicity  $\Delta = 135 \text{ cm}^{-1}$ , or 4.1% of the fundamental transition, in agreement and other C-H stretch modes in diamond [11,12]. With no significant elongation along the diagonal, the  $3323 \text{ cm}^{-1}$  mode’s linewidth of  $9.0 \text{ cm}^{-1}$  (relatively large compared to other C-H stretch modes in diamond [11]) results from a short vibrational lifetime rather than inhomogeneous broadening.

Surprisingly, substantial positive signal was observed at  $3329\text{ cm}^{-1}$ , which increased in intensity after zero time delay, up to a maximum at approximately 2 ps [Fig. 2(f)]. We assign this to a HGS because (i) the signal is above the GSB energy, ruling out ESA features, which are at lower energies due to anharmonicity [11]; (ii) no on-diagonal GSB signal exists at  $3329\text{ cm}^{-1}$ , eliminating contributions from other defects; (iii) the  $N_s\text{:H-C}^0$  bend mode (predicted at  $1400\text{ cm}^{-1}$  [22]) and its harmonics are far removed; and (iv) no off-diagonal cross peaks with  $NVH^0$  are seen [Figs. 2(c) and 2(e)], confirming the signal is isolated to the  $N_s\text{:H-C}^0$  defect.

To explore the vibrational dynamics of the  $N_s\text{:H-C}^0$  defect, we conducted pump-probe spectroscopy at room temperature with a  $3323\text{ cm}^{-1}$  pump and dual probe beams centered at  $3323$  and  $3188\text{ cm}^{-1}$  (Fig. 3). The  $n = 1 \rightarrow 2$  ESA, evident as the red area in Fig. 3(a), begins at time zero (after coherent artifacts within 0.3 ps of time zero) and decays rapidly. In contrast, the HGS feature grows in over the first 2 ps, as shown by the red area in Fig. 3(b) around  $3329\text{ cm}^{-1}$  probe, above the  $n = 0 \rightarrow 1$  GSB (blue).

The transient absorption spectra were modeled by solving the coupled rate equations describing the populations  $N_1$ ,  $N_0$ , and  $N_{0,h}$  of a three-level system [10]. The excited state  $|1\rangle$  can relax directly to the cold ground state

(CGS)  $|0\rangle$  with rate  $1/\tau_d$ , accounting for relaxation processes that leave the defect unperturbed. Alternatively, relaxation can be via an intermediate HGS,  $|0\rangle_h$ , with rate  $1/\tau_h$ , before relaxing to the CGS with cooling rate  $1/\tau_l$ , corresponding to energy transfer away from the defect's local environment. The simulated dynamics [Fig. 3(c)] are in excellent agreement with experiment [Fig. 3(b)], evident in Figs. 3(d)–3(e) by comparing the modeled transient OD (solid lines) with experiment (points) and further by the transient spectra in Fig. 3(f). The comparable lifetimes  $\tau_d = 1.8 \pm 0.2\text{ ps}$  and  $\tau_h = 2.1 \pm 0.2\text{ ps}$  indicate that a substantial fraction of the  $N_s\text{:H-C}^0$  defects are perturbed by a nearby hot phonon distribution during relaxation, while the HGS lifetime  $\tau_l = 3.1 \pm 0.2\text{ ps}$  yielded a slower cooling rate than HGS formation rate, a necessary condition for the observation of a HGS.

Also prominent in Fig. 3(b) are perturbed free induction decay (PFID) oscillations at negative pump-probe delays, involving the  $n = 0$  and  $n = 1$  states [23]. An exponential rise was fit to the  $3323\text{ cm}^{-1}$  PFID signal [data at negative time delays in Fig. 3(e)] yielding the dephasing time of the  $n = 0 \rightarrow 1$  transition,  $T_2 = 1.2\text{ ps}$ . This corresponds to a broader line shape than what was observed for the  $3323\text{ cm}^{-1}$  GSB, suggesting that the HGS may also affect PFID dynamics.

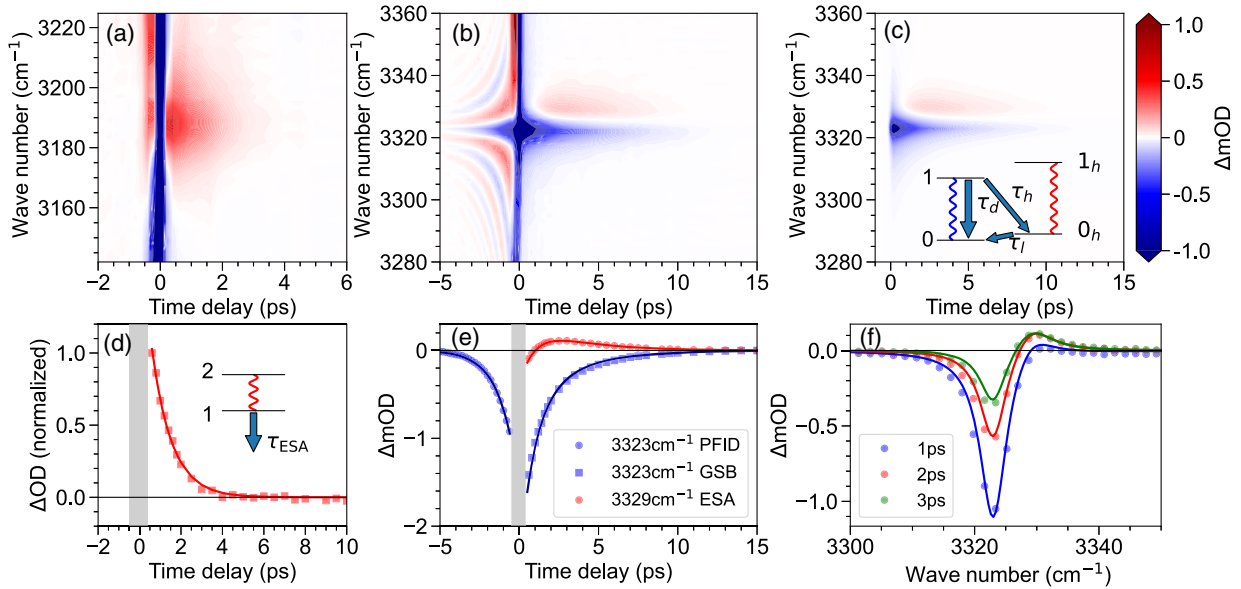


FIG. 3. (a)–(c) Transient optical density maps versus pump-probe delay and wave number from experiment (a), (b) and model (c). (a)  $3323\text{ cm}^{-1}$  pump,  $3188\text{ cm}^{-1}$  probe shows an ESA feature centered at  $3188\text{ cm}^{-1}$  (red,  $t > 0.5\text{ ps}$ ). (b)  $3323\text{ cm}^{-1}$  pump,  $3323\text{ cm}^{-1}$  probe revealed PFID ( $t < 0$ ) centered at  $3323\text{ cm}^{-1}$  that evolved into a GSB for  $t > 0$  (blue). The red positive transient absorption around  $3329\text{ cm}^{-1}$  is assigned to absorption from a HGS. (c) Simulated transient absorption corresponding to experimental data in (b), with  $\tau_d = 1.8\text{ ps}$ ,  $\tau_h = 2.1\text{ ps}$ , and  $\tau_l = 3.1\text{ ps}$ . (d) The normalized integrated signal of the  $3188\text{ cm}^{-1}$  ESA (red squares), fitted with an exponential decay (red line) starting at 0.5 ps and yielding  $\tau_{\text{ESA}} = 0.97\text{ ps}$ . (e) Slices of the experimental data in (b) at  $3323\text{ cm}^{-1}$  for  $t < -0.5\text{ ps}$  (blue circles) are fit by an exponential rise with  $T_2 = 1.2\text{ ps}$ . Experimental data tracking the cold defects (blue squares) and defects with a HGS (red circles) are fit by the solid lines, which are slices through the simulation in (c). (f) Transient optical density spectra from experiment (points) and simulation (lines) at different time delays.

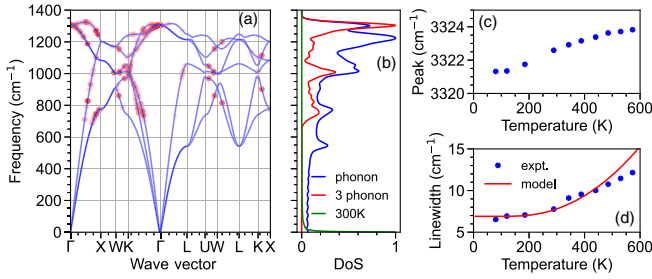


FIG. 4. (a) Phonon dispersion for diamond (blue lines) along with specific phonon modes (semi-transparent red circles) produced by three-phonon emission when the  $N_s\text{:H-C}^0$  defect's  $n = 1$  LVM relaxes. A darker red indicates a greater number of phonon modes involved in HGS generation. (b) Normalized density of states versus frequency for diamond's phonons (blue). The density of nonequilibrium phonon states after LVM relaxation (red) has multiple peaks at high frequencies, while the occupied equilibrium phonon population at room temperature (green) peaks at zero wave number. (c) Temperature dependence of the stretch mode's centre frequency in equilibrium. (d) Experimental temperature dependence of the linewidth (points) and the calculated linewidth (solid line).

The origin of the HGS can be identified by considering the possible phonons produced. A localized lattice distortion produced by phonons [Fig. 1(a)] can alter the LVM bond strength and requires phonons with a short wavelength, corresponding to a high crystal momentum  $\mathbf{k}$ , rather than long-wavelength phonons near the Brillouin zone (BZ) center ( $\Gamma$  point). Phonons with large  $\mathbf{k}$  are a required part of the relaxation process, as evidenced by considering the possible phonons that can be produced such that energy and  $\mathbf{k}$  are conserved. We examined the phonon dispersion curve of diamond [Fig. 4(a)] calculated using density functional perturbation theory [14], which predicts an optical phonon energy of  $1306 \text{ cm}^{-1}$  at the BZ center, within 2% of the experimental value of  $1332 \text{ cm}^{-1}$ . As this is well below the LVM frequency, energy conservation dictates that relaxation from the  $n = 1$  ground state requires the emission of three or more phonons, at least two having appreciable  $\mathbf{k}$ . This results from the fact that no three-particle emission process satisfies energy and momentum conservation using only BZ center ( $\Gamma$ -point) phonons: For example, if two  $\Gamma$ -point phonons from the optical branch ( $1332 \text{ cm}^{-1}$ ) are used, the remaining phonon needs a wave number of  $659 \text{ cm}^{-1}$  and cannot be at the  $\Gamma$  point.

We computationally determined the possible combinations of three phonon emission that can conserve energy and crystal momentum and plot these phonons as semi-transparent red points in Fig. 4(a). The phonons generated have a range of wave vectors across the BZ, primarily along the  $\Gamma$ -X and  $\Gamma$ -K paths, with a distribution in energy pictured in Fig. 4(b) (red line) that can be compared to the phonon density of states (blue line). There are no hot phonons below  $670 \text{ cm}^{-1}$ , and, thus, the lower-energy

acoustic bands are unpopulated, while momentum conservation dictates that some phonons do not contribute to LVM decay (e.g., phonons near the  $L$  point at  $1200 \text{ cm}^{-1}$  do not contribute). The nonequilibrium phonon distribution is dramatically different to the distribution of phonons in thermal equilibrium at room temperature (green line), which peaked toward zero frequency because of the Bose-Einstein factor. Thus, nonequilibrium phonons are generated with a distribution of group velocities: For example, optical phonons around  $1300 \text{ cm}^{-1}$  will have low velocity, as the gradient of the phonon dispersion is low [26]. Such phonons will retain energy in the surroundings of the defect, creating the HGS. In contrast, phonons generated with higher velocity (e.g., in the acoustic branches from  $700$  to  $950 \text{ cm}^{-1}$ ) will remove energy more rapidly.

The efficiency of heat removal from a molecular defect for a particular phonon can be estimated from its mean free path,  $\lambda = v\tau$ , where  $v$  is the phonon speed and  $\tau$  is the time before the phonon scatters. An average  $\tau$  for diamond can be estimated via kinetic theory, which predicts that  $\kappa = \frac{1}{3}C_V v^2 \tau$ , where the heat capacity at constant volume is  $C_V = 1.78 \times 10^6 \text{ J m}^{-3} \text{ K}^{-1}$  for diamond at room temperature. From the phonon dispersion of diamond, longitudinal acoustic phonons near the BZ center have the highest group velocity  $v_{LA} \approx 1.8 \times 10^4 \text{ ms}^{-1}$  and, thus, dominate heat transport in equilibrium. Substituting this into  $\tau = 3\kappa/C_V v^2$  yields  $\tau \approx 11 \text{ ps}$ . Hence,  $\Gamma$ -point LA phonons, on average, travel a distance of  $\lambda = v\tau \approx 200 \text{ nm}$  and can efficiently transfer heat. However, optical phonons and BZ boundary acoustic phonons have shorter scattering times ( $< 1 \text{ ps}$  [26]) and lower group velocities (e.g., around  $10^3 \text{ ms}^{-1}$ ) and, therefore, do not transport heat as effectively, with  $\lambda < 1 \text{ nm}$ . Phonons with small  $\lambda$  will strain the lattice surrounding the defect, creating the HGS.

Uniaxial stress studies on the  $N_s\text{:H-C}^0$  defect show a blueshift of the  $3323 \text{ cm}^{-1}$  feature with increased stress applied to the crystal lattice [7,8,10]. The compression of the defect from uniaxial stress is analogous to the compression provided by the expanded lattice as it becomes populated with phonons, further substantiating our HGS theory. Figure 4(c) shows temperature-dependent absorption in thermal equilibrium [10]. The  $3323 \text{ cm}^{-1}$  feature blueshifts as temperature increases, suggesting that a higher phonon population enhances the energy difference between  $n = 0$  and  $n = 1$ , as corroborated by the  $3329 \text{ cm}^{-1}$  HGS. Commonly, LVMs redshift in frequency at higher temperatures as a result of weaker bonds from lattice expansion, although hydrogen defects in ZnO demonstrate both redshifting and blueshifting with temperature [27], suggesting that defect-lattice coupling leads to nontrivial changes in bond strength (and mode frequency) with temperature.

The  $3323 \text{ cm}^{-1}$  linewidth also increased with temperature [Fig. 4(d)], reflecting a shorter vibrational lifetime for

$n = 1$  when more phonons become available to mediate relaxation. We describe the temperature-dependent linewidth using a model of the interaction between a LVM and a surrounding crystal via three-phonon emission [15], shown by the solid line [10], which agreed with experimental data.

Finally, the HGS recovered to the CGS with a time constant  $\tau_l = 3.1$  ps, corresponding to the timescale required to remove energy from the defect's local environment. The rapid relaxation of the initial nonequilibrium phonons into lower-energy phonons with larger  $\lambda$  dictates  $\tau_l$ : Optical phonons decay via the Klemens channel to produce two acoustic phonons [29,30]. In type IIa single crystal diamond, optical phonons have lifetimes of  $T_{2(\text{phonon})} = 6.9$  ps and  $T_{1(\text{phonon})} = 3.6$  ps [28,29,31,32], with the similarity between  $T_{1(\text{phonon})}$  and  $\tau_l$  further validating our results.

This Letter showed that vibrational relaxation in diamond defects generates localized nonequilibrium phonons that persist long enough to distort the surrounding crystal lattice and the defects' potential energy surface. This challenges the common assumption of a rigid lattice during defect relaxation and establishes HGSs as a key consideration during optical manipulation of functional defects in diamond and other materials. While this Letter was demonstrated on an IR-active, H-related vibrational state rather than a spin state, it remains highly relevant for the quantum information community as spin-phonon coupling is a significant source of spin decoherence in NV and SiV defects. The transient distortion of the local lattice can induce mechanical stress on the defect, which acts as an effective external magnetic field perturbing the spin-state Hamiltonian and thereby affecting their performance as qubits in optically driven quantum information systems [33–35]. This opens an avenue for the manipulation of spin states via local phonon generation. Alternatively, avoiding such effects may also inform the optimisation of spin decoherence times. In addition, our demonstration of localized heating and distortion of the diamond lattice via optical excitation suggests the potential for targeted defect annealing, allowing optimized control of laser writing as a high-precision fabrication technique [36]. Future work should investigate (i) HGS relaxation dynamics at different temperatures, particularly as measurements in equilibrium have demonstrated a nontrivial temperature dependence, and (ii) whether similar defect-lattice interactions occur in other host crystals and defect classes relevant to quantum technologies.

**Acknowledgments**—The 2DIR and transient absorption measurements were carried out using the LIFETIME system at the ULTRA facility within the Science and Technology Facilities Council's Central Laser Facility (United Kingdom), under Application No. 23230009. The authors acknowledge funding from the Engineering and Physical

Sciences Research Council (United Kingdom) under Grant No. EP/V056778/1.

**Data availability**—The data that support the findings of this Letter are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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## End Matter

BC hydrogen has been studied in crystalline and amorphous silicon [37–44], amorphous oxide semiconductors [45], hexagonal boron nitride [46], and gallium nitride [47,48]. In diamond, BC interstitial hydrogen has been studied only theoretically [22,49,50]. Although there

are no experimental data identifying isolated interstitial hydrogen, BC muonium has been identified in diamond [51,52]. Large calculated lattice distortions around BC hydrogen suggest its instability [53], further motivating the use of stable  $N_s:H-C^0$  as a proxy.