

Ultrafast Modulation of Electron-Phonon Couplings and Phonon-Mediated Superconductivity

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Ultrafast modulation of electron-phonon couplings via laser excitation has been experimentally demonstrated, yet a clear microscopic understanding remains elusive. Here, we establish the first *ab initio* framework combining time-dependent density-functional theory, density-functional perturbation theory, and Migdal-Eliashberg theory to track electron-phonon couplings and superconductivity in prototypical phonon-mediated superconductor MgB₂ under laser illumination. We find that near-infrared laser pulses induce charge transfer from boron σ to π orbitals, effectively renormalizing the electron-phonon couplings and boosting the superconducting critical temperature to ~ 52 K. The simulated thermalized superconducting gaps match well with experimental measurements. We propose a generalizable route for ultrafast modulation of superconductivity in materials whose dominant electron-phonon coupling channel can be selectively addressed by light, with photoinduced changes in electronic coupling and screening playing a central role. This Letter offers fundamental insight into nonequilibrium optical control of electron-phonon interactions and superconductivity, and provides a predictive tool for studying photo-modulated electron-phonon coupling and macroscopically observable superconducting properties.

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Electron-phonon coupling (EPC) is a fundamental phenomenon in condensed matter physics. It plays a central role in determining a wide range of physical properties and emergent phenomena [1–17]. The ability to modulate EPC offers a pathway to control material properties and access novel physical states that are otherwise inaccessible under regular conditions. In particular, ultrafast laser excitation offers a powerful, nonthermal route to manipulate electron-phonon coupling on femtosecond timescales. The ultrafast modulation of EPC is a cornerstone for understanding a host of nonequilibrium phenomena where it plays a decisive role, including photoinduced phase transitions [18–21] and transient enhancement of superconductivity [22–25].

Ultrafast modulation of EPC has been experimentally demonstrated through techniques such as terahertz time-domain spectroscopy and time- and angle-resolved photoemission spectroscopy [26–29]. However, the microscopic mechanism through which ultrafast lasers modulate EPC remains elusive. This gap in understanding stems from significant experimental challenges, as directly measuring contributing factors of EPC on ultrafast timescales exceeds the limits of current time and energy resolution capabilities

[30–33]. Concurrently, although real-time time-dependent density-functional theory (TDDFT) has been used to monitor laser-induced changes in selected EPC-related quantities, a complete first-principles framework, which constructs photoexcited-state, $\mathbf{k}$ - and $\mathbf{q}$ -resolved EPC matrix elements through density-functional perturbation theory (DFPT) and applies them directly to anisotropic Migdal-Eliashberg calculations, has not been established [34–36].

Here, we combine TDDFT, DFPT, and Migdal-Eliashberg (ME) formalism [see Sec. S1 of Supplemental Material (SM) [37]] to investigate, from first principles, the photoinduced modulation of electron-phonon coupling and phonon-mediated superconductivity in magnesium diboride (MgB₂). Our *ab initio* calculations reveal that ultrafast modulation of electron-phonon coupling arises from charge transfer excitation between boron σ and π orbitals, leading to an enhanced superconducting critical temperature above 50 K immediately following near-infrared laser excitation. We propose a generalizable route for materials with selectively addressable EPC channels through which ultrafast lasers modulate electron-phonon coupling and phonon-mediated superconductivity (Fig. 1): direct electron excitation introduces nonthermal electron occupation and charge density redistribution, which alter electronic coupling and screening effects, ultimately modulating both EPC and superconductivity.

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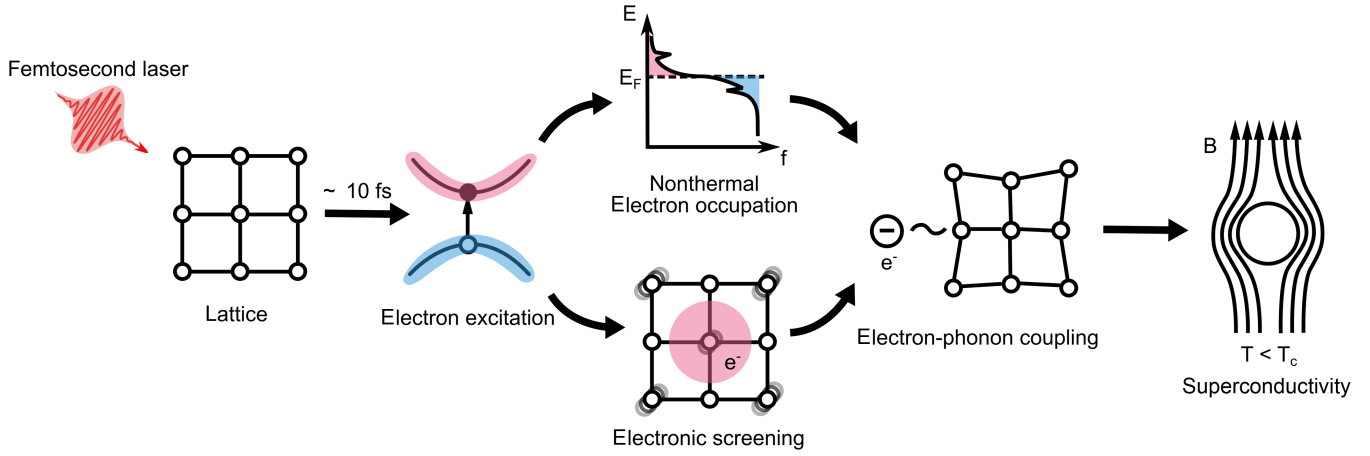


FIG. 1. Schematic of a generalized route via which ultrafast lasers modulate electron-phonon coupling and phonon-mediated superconductivity. After a femtosecond laser pulse is applied, electrons are excited from the valence bands to the conduction bands on the timescale of ~ 10 fs. Nonthermal electron occupation occurs and changes the electronic coupling between different electronic states. Meanwhile, the electronic screening effect is modified due to redistribution of charge density in real space. As a result, the EPC matrix elements and phonon frequencies are renormalized, leading to the modulation of EPC and thus the phonon-mediated superconductivity.

MgB₂ is a well-known BCS superconductor with a critical temperature of $T_c = 39$ K [57] and anisotropic two-band superconducting gaps [58–61], corresponding to boron σ and π orbitals near the Fermi level. The E_{2g} phonon mode, which involves in-plane stretching vibrations of boron atoms, couples most strongly to the σ orbitals and accounts for the majority of the electron-phonon coupling strength [59,62–65]. Thus, the key factor of modulating EPC in MgB₂ is the tunable excitation of σ electrons.

We then apply a Gaussian-type laser pulse polarized along the in-plane zigzag direction of MgB₂ in our simulations (see Sec. S2 of SM [37] for detailed computational parameters). Figures 2(a) and 2(b) show the differential charge density between the ground state ($t < 0$ fs) and the photoexcited state immediately after the laser pulse ($t = 30$ fs). The laser pulse induces a charge transfer from the in-plane boron hexagon (associated with σ orbitals) to the out-of-plane boron π orbitals. This excitation corresponds to electronic transitions from the hole pocket around the Γ point near the Fermi level [Fig. 2(c)]. We note that the excited state is directly related to the nonthermal distribution of photo-carriers, which barely changes at $t = 300$ fs as shown in the right panel of Fig. 2(c). Thus, although the results are obtained for the system at $t = 30$ fs, the laser-induced state in our calculation can be regarded as quasisteady within the lifetime of carrier relaxation (1–100 ps), which is consistent with the lifetime of possible photoinduced superconducting states reported in recent studies [22–24].

The photoexcited charge density $\rho(t)$ will greatly affect the phonon dynamical matrix and the electron scattering potential ($V[\rho]$), which in turn allow us to determine the phonon spectrum and the EPC matrix elements for the excited state (see Secs. S1 and S2 of SM [37] for details) within the framework of perturbation using DFPT [40],

$$g_{nm\nu}(\mathbf{k}, \mathbf{q}, t) = \langle \psi_{m\mathbf{k}+\mathbf{q}}(t) | \frac{\partial V[\rho(t)]}{\partial u_{\mathbf{q}\nu}} | \psi_{n\mathbf{k}}(t) \rangle, \quad (1)$$

where $g_{nm\nu}(\mathbf{k}, \mathbf{q}, t)$ is the EPC matrix element of the excited state, which describes the possibility of scattering an electron from state $|n\mathbf{k}\rangle$ to state $|m\mathbf{k} + \mathbf{q}\rangle$ via the ν th phonon mode with wave vector $\mathbf{q}$ at t . $V[\rho(t)]$ is the time-dependent scattering potential felt by electrons, $u_{\mathbf{q}\nu}$ is the atomic displacement, and $\psi_{n\mathbf{k}}(t)$ and $\psi_{m\mathbf{k}+\mathbf{q}}(t)$ are the time-dependent electron wave functions. The variation of scattering potential induced by E_{2g} in the boron plane is shown in Fig. 2(d). The photoinduced modulation of the variation of scattering potential maintains the inversion symmetry of E_{2g} [Fig. 2(e)] and should directly change the EPC matrix elements upon the laser-induced redistribution of charge density.

At both the ground and excited states, the EPC matrix elements can be monitored with the wave-vector- and phonon-mode-resolved EPC strength $\lambda_{\mathbf{q}\nu}$ and phonon linewidth $\Gamma_{\mathbf{q}\nu}$ as

$$\lambda_{\mathbf{q}\nu} = \frac{\Gamma_{\mathbf{q}\nu}}{2\pi N(\epsilon_F) \omega_{\mathbf{q}\nu}^2}, \quad (2)$$

$$\Gamma_{\mathbf{q}\nu} = 2\pi \omega_{\mathbf{q}\nu} \sum_{nm} \int_{BZ} \frac{d\mathbf{k}}{\Omega_{BZ}} |g_{nm\nu}(\mathbf{k}, \mathbf{q})|^2 \times \delta(\epsilon_{n\mathbf{k}} - \epsilon_F) \delta(\epsilon_{m\mathbf{k}+\mathbf{q}} - \epsilon_F). \quad (3)$$

Here, $N(\epsilon_F)$ is the electronic density of states at the Fermi level ϵ_F and $\omega_{\mathbf{q}\nu}$ is the phonon frequency. $\epsilon_{n\mathbf{k}}$ and $\epsilon_{m\mathbf{k}+\mathbf{q}}$ are the Kohn-Sham energy levels of electronic states $|n\mathbf{k}\rangle$ and $|m\mathbf{k} + \mathbf{q}\rangle$, respectively. Ω_{BZ} is the volume of the Brillouin zone and the integration of $\mathbf{k}$ in Eq. (3) is performed over

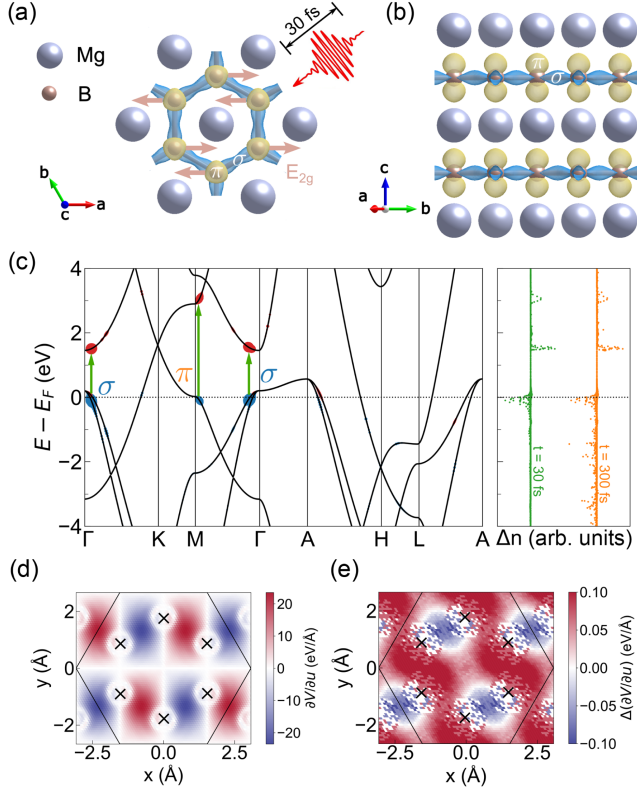


FIG. 2. Top (a) and side (b) view of the atomic structure of MgB_2 and the differential electron density between the ground state and the laser-excited state, which suggests charge transfer excitation from the boron σ to π orbitals. The honeycomb boron layers are intercalated with triangular magnesium layers and each magnesium atom is located at the center of a boron hexagon along the c axis. Regions where electron density increases and decreases are denoted by yellow and blue clouds respectively with an isosurface of 1.5×10^{-5} electron/bohr ^{3} . The light brown arrows in (a) denote the vibration mode of the E_{2g} phonon. (c) The calculated band structure with main electron excitations suggested by the green arrows and the differential electron occupation $[\Delta n(t) = n(t) - n(0)]$ at $t = 30$ fs (green) and $t = 300$ fs (orange), respectively. The blue and red areas are where reduction and increase of the electron occupation occur, respectively, with the size of the colored areas reflecting the amount of change in the electron occupation. (d) The variation of scattering potential at the boron plane originating from atomic displacements of the E_{2g} mode at the ground state. (e) The differential variation of scattering potential of E_{2g} at the boron plane between the excited state and the ground state. The locations of boron atoms are marked with “cross” symbols, and the hexagon denotes the border of the same region shown in (a).

the whole Brillouin zone. As shown in Fig. 3(a), the Γ -A segment of the E_{2g} mode accounts for the majority of the total EPC strength in MgB_2 . Upon laser excitation, the E_{2g} phonon softens along Γ -A due to the reduced σ -electron density in the boron hexagon and weakened electronic screening, as suggested in Fig. 3(b). Both $\Gamma_{\mathbf{q}\nu}$ and $\lambda_{\mathbf{q}\nu}$ increase along Γ -A' while decreasing along A'-A

[Fig. 3(c)], explicitly confirming that the $g_{nm\nu}(\mathbf{k}, \mathbf{q})$ matrix elements associated with E_{2g} are renormalized upon laser excitation.

The Eliashberg spectral function $\alpha^2 F(\omega)$ and cumulative $\lambda(\omega)$ [Fig. 3(d)] show enhancements within the 60–70 meV phonon energy range, which coincides precisely with the energy of the E_{2g} mode along the Γ -A direction. This leads to an overall enhancement of the EPC in MgB_2 and thus the superconducting T_c . The total EPC strength $\lambda = \sum_{\mathbf{q}\nu} \lambda_{\mathbf{q}\nu}$ increases from 0.632 in the ground state, which is consistent with experimental values of $\lambda \approx 0.6$ [60,66], to 0.699 in the excited state [Fig. 3(d)].

We further analyze the electronic origin of the laser-induced enhancement of the EPC by computing the EPC strength contributed by the electronic state $|n\mathbf{k}\rangle$,

$$\lambda_{n\mathbf{k}} = 2 \sum_{m\nu} \int_{BZ} \frac{d\mathbf{q}}{\Omega_{BZ}} \frac{|g_{nm\nu}(\mathbf{k}, \mathbf{q})|^2}{\omega_{\mathbf{q}\nu}} \delta(\epsilon_{m\mathbf{k}+\mathbf{q}} - \epsilon_F), \quad (4)$$

for both the ground and excited states. As shown in Fig. 4(a), the distribution of $\lambda_{n\mathbf{k}}$ exhibits two distinct groups of peaks corresponding to the π and σ superconducting gaps, with the latter exhibiting stronger EPC, which is consistent with previous studies [61,68,69]. The laser pulse affects the two gaps differently: the π gap remains almost unchanged, while the σ gap shifts toward higher $\lambda_{n\mathbf{k}}$. This suggests that only EPC matrix elements involving initial σ orbitals are significantly modulated by the laser. This observation aligns with the laser-induced reduction in electron occupation of σ orbitals near the Fermi level [Fig. 2(c)], which alters the electronic coupling of σ orbitals with nearby states (as illustrated in Fig. 1) and thereby modifies the corresponding EPC matrix elements.

The stronger coupling between σ orbitals and the E_{2g} phonon mode results in a larger superconducting gap, as clearly seen in the temperature-dependent distributions of the anisotropic gap $\Delta_{n\mathbf{k}}$ in Fig. 4(b). In the ground state, the σ gap ranges from 6.3 to 7.5 meV, while the π gap ranges from 1.3 to 2.9 meV, in agreement with previous measurements at 10 K [60,69,70]. After photoexcitation, the σ gap increases to 8.0–9.2 meV, while the π gap shows only a modest increase to 1.6–3.3 meV.

To visualize these changes, we plot the distributions of the superconducting gaps on the Fermi surface at 10 K for the ground state [Fig. 4(c)] and the difference between the excited and ground states [Fig. 4(d)]. Consistent with Fig. 4(b), the π gap shows negligible change, while the σ gap increases noticeably. Furthermore, the average σ -gap magnitude at 0 K, estimated from the quasiparticle density of states, increases from 7.3 to 9.0 meV, while the π gap increases only slightly from 2.9 to 3.2 meV (see Fig. S9 in SM [37]), in line with the above analysis. These enhancements lead to an increase in the superconducting transition temperature T_c . In the ground state, the calculated T_c is approximately 40 K, matching the experimental value of

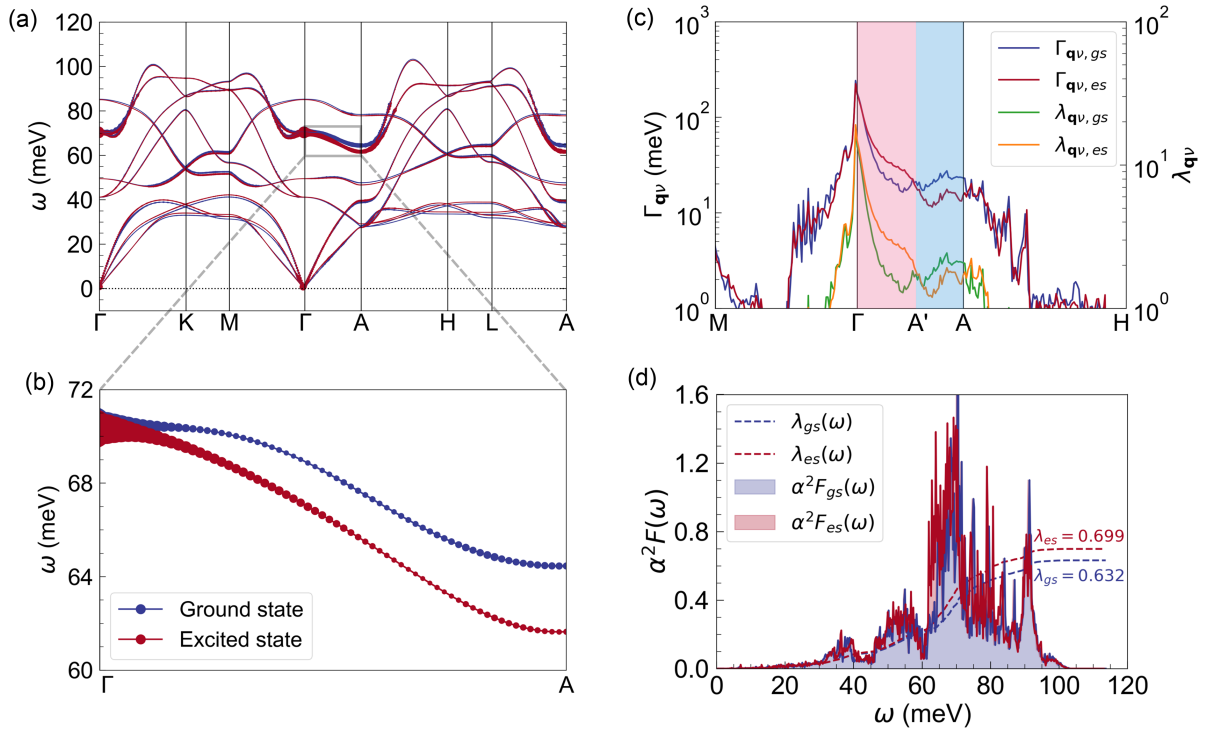


FIG. 3. (a) The $\lambda_{\mathbf{q}\nu}$ -distinguished phonon spectrum of MgB_2 at the ground state (blue) and the excited state (red). The size of the dots along the phonon dispersion reflects the magnitude of $\lambda_{\mathbf{q}\nu}$. (b) Enlargement of the region inside the gray rectangle in (a), which shows the phonon dispersion and $\lambda_{\mathbf{q}\nu}$ of the E_{2g} mode along Γ -A. (c) Phonon linewidth $\Gamma_{\mathbf{q}\nu}$ and $\lambda_{\mathbf{q}\nu}$ of the E_{2g} mode at the ground state ($\Gamma_{\mathbf{q}\nu,gs}$, $\lambda_{\mathbf{q}\nu,gs}$) and the excited state ($\Gamma_{\mathbf{q}\nu,es}$, $\lambda_{\mathbf{q}\nu,es}$) along the high-symmetry path in the reciprocal space. A' is a point defined on the path of Γ -A, which divides Γ -A into two areas in light red and blue, respectively. (d) Eliashberg spectral function $\alpha^2F(\omega)$ and the cumulative EPC strength $\lambda(\omega)$ at the ground state [$\alpha^2F_{gs}(\omega)$, $\lambda_{gs}(\omega)$] and the excited state [$\alpha^2F_{es}(\omega)$, $\lambda_{es}(\omega)$].

39 K [57]. For the photoexcited state, the T_c derived from the superconducting gap Δ_{nk} reaches 52 K [Fig. 4(b)], indicating a photoinduced enhancement of about 12 K.

Discussion—We first examine the key approximations underlying the above superconductivity calculations. The laser-induced temperature rise in our simulations is at most 0.13 K (see Sec. S6 of SM [37]), which is negligible compared to the T_c of MgB_2 and rules out significant thermal effects on the calculated T_c . Although the laser pulse produces a nonthermal carrier distribution [Fig. 2(c)], the occupation perturbation within the near-Fermi-level electronic subspace sampled by the ME Fermi-surface delta-function kernel remains very small under the present weak-excitation conditions, validating the quasistatic ME framework as a controlled approximation to the exact pairing kernel for this regime (see Sec. S8 of SM [37]). In addition, because the laser-modified screening may in principle renormalize the Coulomb pseudopotential, we repeated the photoexcited-state ME analysis over the empirical range $\mu_c^* = 0.12$ –0.20. The enhancing trend persists throughout this range, confirming that the laser-induced enhancement of superconductivity in MgB_2 is robust for the ultrafast time window and weak-excitation laser parameters considered here (see Sec. S9 of SM [37]).

We next distinguish this nonthermal femtosecond response from polarization and thermalized-response effects. When the laser pulse is polarized out of plane, the superconductivity remains nearly unchanged (see SM Sec. S3 [37]), demonstrating anisotropic responses of the material to lights with different polarization directions. Previous pump-probe experiments [71–73] have shown the “hot phonon” physics in MgB_2 , i.e., the initial relaxation of photoexcited electrons proceeds by generating a large population of the optical E_{2g} phonon, which breaks Cooper pairs below T_c on picosecond to nanosecond timescales [71,72]. In a recent ultrafast experiment by Sobolev *et al.*, fluence-dependent gap suppression is observed in MgB_2 after near-infrared excitation and electron thermalization [67]. These phenomena suggest that the initial nonthermal electron occupation would be crucial to the ultrafast modulation of EPC and superconductivity in our calculations. The earlier few-hundred-femtosecond regime has not yet been clearly isolated in published MgB_2 experiments, but should in principle be accessible with sufficiently fast time-resolved probes before hot-phonon thermalization dominates. To verify the distinction between nonthermal and thermalized responses, we calculate the superconducting gaps of the ground state under

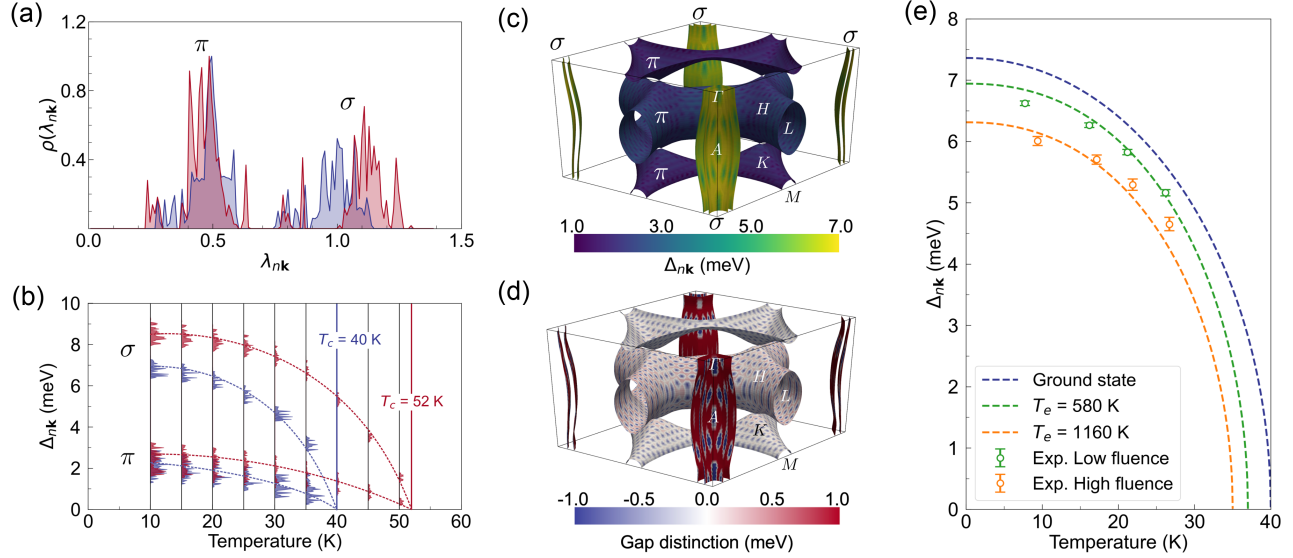


FIG. 4. (a) The distribution of the state- and wave-vector-resolved EPC strength λ_{nk} at the ground state (blue-shaded area) and at the excited state (red-shaded area). (b) The anisotropic superconducting gaps Δ_{nk} at different temperatures at the ground state (blue) and the excited state (red). The Coulomb potential μ_c^* is set to 0.16. The dashed lines are references for the eye. The superconducting critical temperature T_c is determined as the temperature at which the superconducting gaps disappear. (c) The ground-state gap-distinguished Fermi surface of MgB_2 at 10 K. The two cylindrical sheets around Γ -A are derived from the boron σ orbitals while the tubular sheets around K-M and H-L are derived from the boron π orbitals. (d) The differential gap-distinguished Fermi surface between the ground state and the excited state at 10 K. Red denotes an increase while blue represents a reduction of the gap value after excitation. Note the regularly distributed dots on both the σ and π sheets in (c) and (d) originate from numerical error in the interpolation of the gaps on the Fermi surface. The topology of the Fermi surface barely changes after excitation. (e) The suppressed average σ gap of the ground state with thermally distributed electrons at higher electronic temperatures. The dashed curves are obtained by fitting the average gap values extracted from the superconducting quasiparticle density of states with $\Delta(T) = \Delta(T=0)[1 - (T/T_c)^p]^{0.5}$. The open circles are adapted from the experimental results of Sobolev *et al.* [67].

thermal electronic distributions with certain effective electronic temperatures. As shown in Fig. 4(e), the amplitude of the σ gap is suppressed as the electronic temperature increases, in clear contrast to the enhanced gap observed in Fig. 4(b) with laser-induced nonthermal electronic distribution [Fig. 2(c)]. Assuming the original gap in the experiment by Sobolev *et al.* [67] follows the temperature dependence of the calculated ground-state gap [blue dashed line in Fig. 4(e)], the gaps suppressed by near-infrared lasers under experimental conditions agree well with the calculated gaps at higher electronic temperatures [Fig. 4(e)]. This indicates that the thermal-equilibrium electronic state in our calculations resembles the thermalized state in experiment. Moreover, the enhancement of EPC and transient phonon-mediated superconductivity at $t = 30$ fs observed in our calculations is dominated by the initial nonthermal electron occupation after laser excitation.

Our TDDFT + DFPT + ME calculations demonstrate a photoinduced enhancement of both the EPC and T_c in MgB_2 under near-infrared laser pumping. Although the electronic energy eigenvalues and density of states near the Fermi level do not change substantially after photoexcitation and the near-Fermi-level occupations sampled by the ME kernel remain close to the Fermi-Dirac profile, the laser pulse creates nonthermal occupations away from the Fermi level and induces a real-space redistribution of

electronic charge involving charge transfer from boron σ to π orbitals. This charge redistribution modifies electronic screening, softens the E_{2g} phonon along Γ -A, and changes the EPC matrix elements. These combined effects increase the total EPC strength λ and promote BCS superconductivity. We propose a generalizable route for ultrafast optical control of EPC and superconductivity in materials with selectively addressable EPC channels (Fig. 1), highlighting the critical role of laser-induced real-space charge redistribution and screening renormalization. Although a full treatment of the far-from-equilibrium regime may require combining ME theory with Floquet theory [74] or non-equilibrium Green's function methods [75,76], our approach offers a promising tool for near-equilibrium and quasi-steady-state conditions, providing an effective strategy for designing superconductivity and other light-controlled quantum properties.

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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