

Enhancement of superconductivity with external phonon squeezing

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Squeezing of phonons due to the nonlinear coupling to electrons is a way to enhance superconductivity as theoretically studied in a recent work [Kennes *et al.*, *Nat. Phys.* **13**, 479 (2017)]. We study quadratic electron-phonon interaction in the presence of phonon pumping and an additional external squeezing. Interference between these two driving sources induces a phase-sensitive enhancement of electron-electron attraction, which we find as a generic mechanism to enhance any boson-mediated interactions. The strongest enhancement of superconductivity is shown to be on the boundary with the dynamical lattice instabilities caused by driving. We propose several experimental platforms to realize our scheme.

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Optical excitation of infrared-active (IR-active) phonon modes allows for ultrafast pumping of solid-state systems into nonequilibrium states exhibiting a wide range of exotic properties [1]. In particular, this enables manipulation of magnetic states [2], charge orders [3], and superconductivity [4,5]. A possible explanation of the transient enhancement observed in [4] is the parametric driving of Raman phonons by IR phonons [6,7]. Enhanced [8] quantum fluctuations of phonon modes lead to a stronger phonon-mediated attraction between electrons, thus enhancing the superconductivity.

Another proposed mechanism of transient superconductivity enhancement is based on a nonlinear coupling between electrons and phonons [9,10]. In this case, the squeezing of phonons is generated directly by the electron-phonon interaction, since the coupling is quadratic in the phonon operator. As shown in [10] due to this nonlinearity, the effective interaction is enhanced proportionally to the coherent excitation rate of the phonon mode.

In this Letter, we clarify the role of squeezing and study the possibility of enhancement of superconductivity by an additional external parametric drive. We consider a model that combines both ingredients—the linear and parametric driving of phonons that are nonlinearly coupled to electrons. In order to illustrate the influence of parametric driving on a bosonic degree of freedom $[a, a^\dagger] = 1$ and introduce a related terminology, we define the Hamiltonian of a parametrically driven harmonic oscillator [11] with a bare frequency ω_0 as $H_{\text{PO}} = \omega_0 \hat{a}^\dagger \hat{a} - D(\hat{a}^2 e^{2i\omega_p t} + \hat{a}^{\dagger 2} e^{-2i\omega_p t})/2$ where D stands for the squeezing strength and ω_p is the parametric driving frequency. One can show that the expectation value of the quadrature $\hat{X}_\theta \equiv \hat{a}e^{i\theta} + i\omega_p t + \text{H.c.}$ decreases for $\theta = \pi$ and increases for $\theta = 0$, with respect to a local oscillator. We refer to these quadratures as “squeezed” and “antisqueezed,” respectively. Similarly, the corresponding retarded correlation function of these quadratures, which determines the strength of mediated interaction by these bosonic modes, can decrease or increase in a phase-sensitive fashion (see the Supple-

mental Material [12] for spins as an illustrative example). Consequently, the squeezing of proper phonon quadrature can significantly amplify the phonon-mediated electron-electron interaction. Moreover, the parametric drive can soften the phonon modes that further amplifies the electronic interaction. In this work, we demonstrate that such amplifications lead to the enhancement of superconductivity, by analytically and numerically employing the Migdal-Eliashberg theory. The superconducting critical temperature T_c is shown in Fig. 1(c) as a function of external phonon squeezing rate D . The external parametric drive that is the crucial element of our proposal can be achieved by either exploiting intrinsic photon-phonon coupling nonlinearities [13] or by using a parametric optical amplifier in an optical cavity to produce squeezed light [14] as schematically shown in Fig. 1(a).

To be specific, we study a two-dimensional superconductor interacting with an infrared-active optical phonon mode and consider the coupling to be quadratic in a phonon operator [4,9,10]. Linear coupling terms can also be present without affecting the results below. The full Hamiltonian of the system reads as $\hat{H}_{\text{full}} = \hat{H}_p + \hat{H}_e + \hat{H}_{e-p} + \hat{V}(t)$:

$$\begin{aligned} \hat{H}_p = & \underbrace{\sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{a}_{\mathbf{q}}^\dagger \hat{a}_{\mathbf{q}}}_{H_p} + \underbrace{\sum_{\mathbf{k}, \sigma} (\epsilon_{\mathbf{k}} - \mu) \hat{c}_{\mathbf{k}, \sigma}^\dagger \hat{c}_{\mathbf{k}, \sigma}}_{H_e} \\ & + \underbrace{\frac{g}{N} \sum_{\sigma, \mathbf{k}, \mathbf{q}, \mathbf{q}'} c_{\mathbf{k}+\mathbf{q}-\mathbf{q}', \sigma}^\dagger \hat{c}_{\mathbf{k}, \sigma} \hat{\phi}_{\mathbf{q}} \hat{\phi}_{-\mathbf{q}'}}_{H_{e-p}}, \end{aligned} \quad (1)$$

where N denotes the total number of lattice sites, $\mathbf{q}$ is the lattice quasimomentum vector, $\omega_{\mathbf{q}}$ and $\epsilon_{\mathbf{k}}$ respectively stand for the phonon and electron dispersions, $\hat{c}_{\mathbf{k}, \sigma}$ is the electron annihilation operator, and $\hat{\phi}_{\mathbf{q}} \equiv \hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^\dagger$ is the phonon displacement field operator. Phonons are linearly and parametrically driven at frequency ω_p with the corresponding driving strengths α and $D_{\mathbf{q}}$. The external driving Hamiltonian

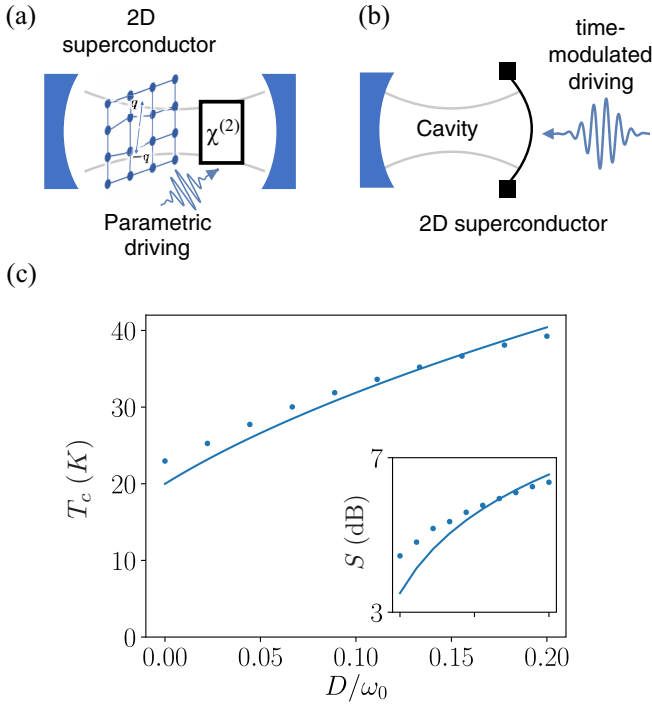


FIG. 1. Steady-state enhancement of superconducting transition temperature T_c . (a), (b) Sketch of the setup: 2D superconductor on an optomechanical membrane. Phonon squeezing is induced by (a) the squeezed light produced by the optical parametric oscillator and (b) time modulation of the classical input light. (c) Superconducting transition temperature as a function of the parametric driving strength D for optimal value of the detuning δ , assuming the phonon driving is fixed at $\alpha = 0.2\omega_0$: parametrically driven phonons (blue). The corresponding phonon excess noise $S \equiv N^{-1} \sum_q \langle \hat{\phi}_q \hat{\phi}_{-q} \rangle$ is shown in the inset. The frequency of the IR-active phonon mode is taken to be $\omega_0 = 0.17$ eV corresponding to the frequency of the infrared-active phonon mode of the K_3C_{60} fulleride superconductor [6,9].

reads

$$\hat{V}(t) = 2\alpha \cos(\omega_p t + \theta_\alpha) \hat{\phi}_0 + \sum_q D_q \cos(2\omega_p t + \theta_D) \hat{\phi}_q \hat{\phi}_{-q}, \quad (2)$$

where $\theta_{\alpha,D}$ are the relative phases of linear and parametric drivings. As we discuss below, these phases allow one to control the strength of coupling to electrons. We note that the model becomes dynamically unstable at strong parametric drive D_q [11]. This is manifested in the exponential growth of the phonon displacement $\langle \hat{\phi}_q \rangle$, as a function of time. Therefore, we impose $\omega_q - \omega_p \geq D_q$ to avoid such an instability.

The external drive induces a finite expectation value of the zero-momentum phonon mode $\langle \hat{\phi}_0 \rangle$. We treat this in terms of mean-field theory and keep quadratic fluctuations. We perform two unitary transformations of the Hamiltonian equation (1). First, we consider the frame, rotating at the phonon driving frequency ω_p , which transforms bosonic variables as $\hat{a}_q \rightarrow \hat{a}_q e^{-i\omega_p t}$. Second, we perform a shift of the zero-momentum bosonic variables $\hat{a}_q \rightarrow \hat{a}_q + \bar{a}_0 \delta_{q,0}$, where $\bar{a}_0$ denotes the adiabatic steady-state coherence to the lowest

order in $1/\omega_p$ (see Supplemental Material):

$$\bar{a}_0 \equiv \alpha \frac{D_0 e^{i(\theta_\alpha - \theta_D)} - \delta_0 e^{-i\theta_\alpha}}{\delta_0^2 - D_0^2}. \quad (3)$$

Finally, we perform the rotating-wave approximation and discard the rotating at frequencies $\propto 2\omega_p$, by assuming that the driving frequency ω_p is the largest energy scale in the system. As shown in the Supplemental Material, the effective coupling in the model is maximized for the following choice of driving phases $\theta_D = \pi$, $\theta_\alpha = 0$. This choice corresponds to antisqueezing of the quadrature to which electrons are coupled. With these approximations and neglecting all nonlinear and rotating contributions, the phonon Hamiltonian and the electron-phonon Hamiltonians are transformed as

$$\hat{H}_{ph} = \sum_q \delta_q \hat{a}_q^\dagger \hat{a}_q - \sum_q \frac{D_q}{2} (\hat{a}_q \hat{a}_{-q} + \hat{a}_q^\dagger \hat{a}_{-q}^\dagger), \quad (4)$$

$$\hat{H}_{int} = \frac{g_{eff}}{\sqrt{N}} \sum_{\sigma,q} \hat{c}_{k+q,\sigma}^\dagger \hat{c}_{k,\sigma} (\hat{a}_q + \hat{a}_{-q}^\dagger), \quad (5)$$

where the detuning $\delta_q \equiv \omega_q - \omega_p$ and the effective electron-phonon coupling is $g_{eff} = 2gl\bar{a}_0/\sqrt{N}$. Equation (4) is equivalent to a nondegenerate multimode parametric oscillator [11] below the parametric instability threshold for $\delta_q \geq D_q$, which can be diagonalized by means of the Bogolyubov transformation $\hat{a}_q = \cosh(r_q) \hat{b}_q + \sinh(r_q) \hat{b}_{-q}^\dagger$ with $\zeta_q = 2^{-1} \text{arctanh}(D_q/\delta_q)$. We find $\hat{H}_{ph} = \sum_q \sqrt{\delta_q^2 - D_q^2} \hat{b}_q^\dagger \hat{b}_q$ and $\hat{H}_{int} = N^{-1/2} g_{eff} \sum_{\sigma,q} e^{\zeta_q} \hat{c}_{k+q,\sigma}^\dagger \hat{c}_{k,\sigma} (\hat{b}_q + \hat{b}_{-q}^\dagger) e^{i\mathbf{q} \cdot \mathbf{r}_i}$. Close to the parametric instability $D_q \sim \delta_q$, the coupling scales as $e^{\zeta_q} \propto (1 - D_q/\delta_q)^{-1/4}$. Equations (4) and (5) are therefore equivalent to a conventional Holstein model [15] with the softened phonons and an enhanced electron-phonon coupling. As we show below, the combination of these factors can lead to an enhanced T_c compared to the configuration without squeezing.

In order to show this enhancement, we consider the squeezed electron-phonon model of Eqs. (4) and (5) within the equilibrium Migdal-Eliashberg (ME) theory [16,17] and provide an estimate of the superconducting phase transition temperature T_c . ME theory relies on the Migdal theorem that allows one to neglect vertex corrections to the electron Green's function provided they are much faster than phonons. In the case of the effective Holstein model (4), (5), this is characterized by $\sqrt{\delta_q^2 - D_q^2} \ll E_F$, where E_F is the Fermi energy. The remaining equations for the electronic and phonon self-energies form a closed set of equations, which can be solved self-consistently, and we consider the formulation of the theory above the critical temperature $T \gtrsim T_c$.

We start by defining the fully renormalized imaginary-time propagators $\mathcal{G}_{\mathbf{k}}^{-1} = i\omega_n - (\epsilon_{\mathbf{k}} - \mu) - \Sigma_{\mathbf{k}}(i\omega_n)$, $\mathcal{D}_{\mathbf{k}}^{-1} = \mathcal{D}_{\mathbf{k}}^{(0)-1}(i\omega_m) - \Pi_{\mathbf{k}}(i\omega_n)$, where the unperturbed squeezed phonon propagator is $\mathcal{D}_{\mathbf{k}}^{(0)}(i\omega_m) = -2(\delta_q + D_q)/(\omega_m^2 + \delta_q^2 - D_q^2)$ and $\omega_n = \pi(2n+1)/\beta$, $\omega_m = 2\pi m/\beta$, $m, n \in \mathbb{Z}$ denote fermionic and bosonic Matsubara frequencies, respectively.

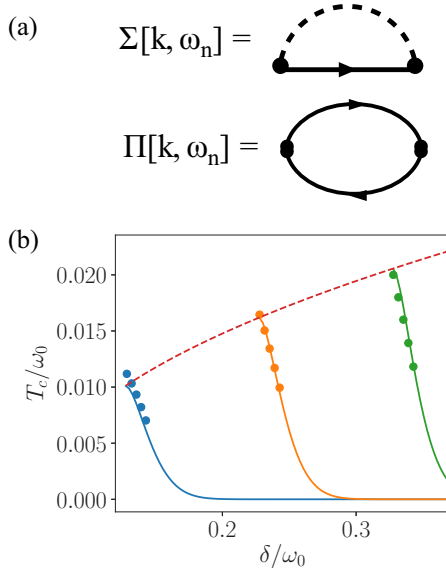


FIG. 2. Enhancement of superconductivity. (a) Electronic and phononic self-energies within Migdal-Eliashberg theory. Solid and dashed lines respectively stand for the fully renormalized electron and phonon propagators. (b) Superconducting T_c as a function of the detuning δ : configuration without squeezing $D = 0$ (blue), $D = 0.1\omega_0$ (orange), and $D = 0.2\omega_0$ (green); and optimal values (red dashed). Dotted curves correspond to the exact numerical solution of the Migdal-Eliashberg equations (6)–(8) on a discretized momentum-frequency lattice.

The electronic and phononic self-energies obey the equations [15], as diagrammatically shown in Fig. 2(a):

$$\Sigma_{\mathbf{k}}(i\omega_n) = -\frac{g_{\text{eff}}^2}{\beta N} \sum_{m, \mathbf{q}} \mathcal{D}_{\mathbf{k}-\mathbf{q}}(i\omega_n - i\omega_m) \mathcal{G}_{\mathbf{q}}(i\omega_m), \quad (6)$$

$$\Pi_{\mathbf{k}}(i\omega_n) = \frac{2g_{\text{eff}}^2}{\beta N} \sum_{m, \mathbf{q}} \mathcal{G}_{\mathbf{q}}(i\omega_m) \mathcal{G}_{\mathbf{q}-\mathbf{k}}(i\omega_m - i\omega_n). \quad (7)$$

These equations define the properties of the normal state of the electron gas. In order to find the superconducting transition temperature, we solve the linearized self-consistent equation for the pairing vertex $\Gamma_{\mathbf{k}}$:

$$\Gamma_{\mathbf{k}}(i\omega_n) = -\frac{g_{\text{eff}}^2}{N\beta} \sum_{\mathbf{q}} \mathcal{D}_{\mathbf{k}-\mathbf{q}}(i\omega_n - i\omega_m) \Gamma_{\mathbf{q}}(i\omega_m) \times \mathcal{G}_{\mathbf{q}}(i\omega_m) \mathcal{G}_{-\mathbf{q}}(-i\omega_m). \quad (8)$$

The highest-temperature solution of this equation defines the critical temperature T_c . We provide an analytical solution of Eqs. (6)–(8) under several simplifying assumptions. In particular, we assume that the detuning $\delta_{\mathbf{q}}$ and the squeezing parameter $D_{\mathbf{q}}$ do not depend on momentum $\mathbf{q}$. In this case, the only momentum dependence in Eqs. (6) and (7) is due to the electron polarization operator. The latter Eq. (7) contains static $\omega_n = 0$ and dynamical contributions $\omega_n \neq 0$. The static contribution is responsible for the phonon softening due to the interaction with electrons and it is generally important at strong couplings. In addition, it effectively enhances the electron-phonon interaction [18]. The dynamical contribution

describes the Landau damping. We neglect the dynamical contribution as it is smaller than the first Matsubara frequency term in the denominator of $\mathcal{D}$ in the relevant temperature ranges [18]. In addition, we restrict the polarization operator in Eq. (7) to its zeroth Matsubara component taken with respect to the unperturbed fermionic Green's function: $\Pi_{\mathbf{k}}(i\omega_n) \approx -2\nu_0 g_{\text{eff}}^2$, where $\nu_0 \equiv N^{-1} \sum_{\mathbf{k}} \delta(E_F - \epsilon_{\mathbf{k}})$ is the density of states at the Fermi energy E_F . We note that according to this definition, ν_0 has the dimension of inverse energy. Under these assumptions the renormalized phonon propagator takes the following form:

$$\mathcal{D}(i\omega_n) = \frac{-2(\delta + D)}{\omega_n^2 + (\delta^2 - D^2)(1 - 2\lambda_0)}, \quad (9)$$

where the effective electron-phonon coupling is defined as $\lambda_0 = 2\nu_0 g_{\text{eff}}^2 / (\delta - D)$. The antisqueezing manifests itself as an excess noise of the phonon field $\hat{\phi}_{\mathbf{q}}$, which is found by taking the Matsubara frequency sum in Eq. (9) in the $T \rightarrow 0$ limit $\langle \hat{\phi}_{\mathbf{q}} \hat{\phi}_{-\mathbf{q}} \rangle = -\beta^{-1} \sum_n \mathcal{D}_{\mathbf{q}}(i\omega_n) \approx \sqrt{(\delta + D)/[(1 - 2\lambda_0)(\delta - D)]}$. We see that the phonon fluctuations are enhanced by interaction with electrons and by external squeezing in a multiplicative way.

Since the right-hand sides of Eqs. (6) and (8) do not depend on momentum $\mathbf{k}$, the dependence can be eliminated by taking an average over Fermi surface $\Gamma_n \rightarrow \langle \Gamma_{\mathbf{k}}(i\omega_n) \rangle_{\text{FS}}$, $\Sigma_n \rightarrow \langle \Sigma_{\mathbf{k}}(i\omega_n) \rangle_{\text{FS}}$. An approximate analytical solution of these equations is known [19,20], and yields the following expression for the critical temperature:

$$T_c = \frac{\sqrt{(\delta^2 - D^2)(1 - 2\lambda_0)}}{1.2} e^{-1.04[(\lambda_{\text{eff}} + 1)/\lambda_{\text{eff}}]}, \quad (10)$$

where the effective coupling strength is defined as $\lambda_{\text{eff}} = \lambda_0 / (1 - 2\lambda_0)$ [18], and the first term in this expression stands for the effective phonon bandwidth, which corresponds to the poles of Eq. (9) with respect to the Matsubara frequency. At strong coupling, λ_0 , the system undergoes a transition to charge-density phase [17,18,21]. In Eq. (10), it manifests itself as singularity of λ_{eff} at $\lambda_0^{\text{cr}} = 0.5$. Due to the vertex corrections neglected in Eqs. (6)–(8), the exact Monte Carlo treatment of the Holstein model [17] predicts a slightly different value $\lambda_0^{\text{cr}} \approx 0.4$. In the following, we will restrict all system parameters such that $\lambda_0 \leq \lambda_0^{\text{cr}}$ in order to avoid this instability.

We now analyze Eq. (10) by varying δ and D while assuming g_{eff} is fixed. In the absence of squeezing ($D = 0$), we find the maximum T_c with respect to the detuning δ being equal to $T_c^{\text{max}} \approx 0.4g_{\text{eff}}^2\nu_0$. This value, being expressed in terms of the optimal detuning, is equal to $T_c^{\text{max}} \approx 0.08\delta$, which reproduces the known result [17,22]. In order to study the influence of squeezing on the superconducting temperature, we assume the squeezing parameter D is fixed to some positive value. In this case a new maximum with respect to δ is straightforwardly found to be $T_c^{\text{max}}(D) \approx 0.25\sqrt{g_{\text{eff}}^2\nu_0 D}$ in the limit when $D \gg g_{\text{eff}}^2\nu_0$. It is achieved at $\delta^{\text{max}} \approx D + 5g_{\text{eff}}^2\nu_0$. This combination of squeezing and detuning saturates the bare electron-phonon coupling to $\lambda_0 \approx \lambda_0^{\text{cr}}$, which is approximately independent of D and δ . The effective phonon bandwidth for the optimal detuning scales as $\sqrt{(\delta^2 - D^2)(1 - 2\lambda_0)} \propto$

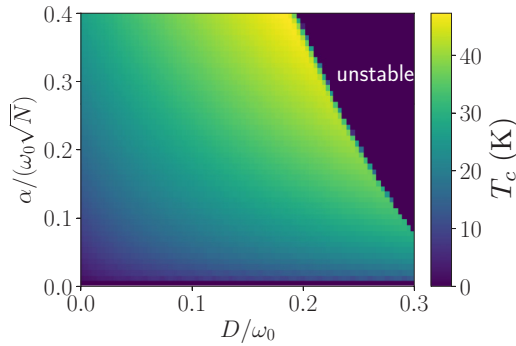


FIG. 3. Superconducting critical temperature T_c as a function of the linear driving α and parametric driving strength D . The detuning is chosen to produce the strongest effect at each point. The strongest enhancement is achieved at the boundary of the lattice instability region.

$\sqrt{g_{\text{eff}}^2 \nu_0 D}$, which determines the scaling of T_c at large squeezing. The enhancement can therefore be seen as increasing the effective bandwidth of phonons while keeping the effective coupling fixed to its maximal value [17,22]. In general, g_{eff} also depends on D and δ due to being proportional to the steady-state phonon occupation. The presented analysis can be straightforwardly extended to take this into account.

We now compare the analytical prediction for the critical temperature with the numerical self-consistent solution of Eqs. (6) and (8) performed in a discretized 52×52 momentum and 400 Matsubara frequency lattice space. We consider the external driving $\alpha = 0.25\omega_0$ to be fixed, while we vary the detuning δ . The critical temperature as a function of the detuning is shown in Fig. 2 for several values of the squeezing parameter D . The smallest detuning of all curves corresponds to $\lambda_0 \approx \lambda_0^{\text{cr}}$. The maximum T_c is achieved at the lowest possible δ in agreement with the analytical expression provided above. We study the effect of linear α and parametric D driving on superconducting T_c for optimal values of detuning δ (Fig. 3). The external squeezing allows one to achieve strong enhancement at much lower driving intensities α , and the strongest effect is achieved on the boundary of the lattice instability regimes.

We now discuss two possible experimental realizations of our idea to generate phonon squeezing. The first proposal exploits the intrinsic photon-phonon coupling nonlinearities [13]. For illustration purposes, we consider a simplified model of two-dimensional electron lattice gas with the nearest-neighbor tunneling rate $J \approx 0.2\omega_0$, corresponding to the K_3C_{60} fulleride superconductor [6,9]. We assume that the infrared-active phonon mode with Debye frequency $\omega_0 \approx 0.17$ eV being driven by a bichromatic light at frequencies ω_p and $2\omega_p$. For an estimate of achievable phonon parametric driving rate, we take the values achieved with phonon parametric amplification [13] $D \propto 0.1\omega_0$ as achieved at field values of the order of 10 MV/cm. Here we focus only on pairing induced by external driving. In our simulations we consider the electron-phonon coupling coefficient $g \approx 0.1\omega_0$ [9] and the electron density of states $\nu_0 \approx 0.6\omega_0^{-1}$. We note that in deriving Hamiltonians (4) and (5) we neglected

the terms rotating at $\propto 2\omega_0$ [23] (see Supplemental Material) which may induce heating for broader-band materials. Our simplified analysis can be extended with taking these rotating terms into account perturbatively [6]. With the parameters above we estimate the bare electron-phonon coupling $g^2 \nu_0 \approx 0.006\omega_0$. The corresponding values of the critical temperature are shown in Fig. 1(c).

In the second approach, we consider a two-dimensional [24] superconductor optomechanical membrane optomechanically coupled to a cavity mode as shown in Figs. 1(a) and 1(b). Experimentally, the cavity optomechanical coupling to the two-dimensional (2D) van der Waals system has recently been demonstrated in [25–27]. The main challenge in this case is to control the high-frequency phonons as the critical temperature, Eq. (10), is proportional to the overall frequency range of the phonon modes. Coupling of light to high-frequency phonons has been demonstrated in several setups including the optomechanical disk resonators [28] and high-frequency bulk acoustic phonons [29]. We consider two possible ways of phonon squeezing as illustrated in Fig. 1. First, squeezing can be achieved via hybridization with photons which are parametrically driven [14,30]. Alternatively, as we show in the Supplemental Material [31], squeezing of membrane can be performed by a very specific time modulation of incoming light. By assuming a frequency range of the order of 100 GHz, as achieved in resonators based on acoustic distributed Bragg reflectors [32–37], we can estimate the achievable enhancement to be of the order of $T_c \propto 3$ K for the same parameter ratio as provided in the previous paragraph. The main limiting factor is the effective phonon bandwidth which is substantially reduced due to squeezing close to the parametric instability. However, the analysis presented in this Letter is restricted to the isotropic case, i.e., when $D_{\mathbf{q}} = D$, $\delta_{\mathbf{q}} = \delta$. The momentum dependence of the $D_{\mathbf{q}}$ and $\delta_{\mathbf{q}}$, which is generally present in experiment, provides an additional degree of freedom. In particular, this allows one to control the effective phonon dispersion independently of the coupling strength. The enhancement of electron interaction may be expected in the case of parametrically driving only the phonons with $q = 2k_F$, where k_F is the Fermi momentum.

In conclusion, we studied the enhancement of superconductivity due to an externally induced squeezing. The phase-sensitive squeezing enhances quadrature fluctuations of the phonon field leading to exponentially stronger interaction, while reducing the spectral bandwidth of phonons. We study the competition of these two effects numerically and analytically and find a parameter range of enhanced superconductivity. The effective squeezed Holstein model describing the system allows one also to dynamically suppress coupling to a certain range of phonon modes. The strength of the suppression is exponential. This can be very useful in the case when superconductivity competes with other types of instabilities, e.g., charge-density wave instability. By decoupling from the phonon modes responsible for the instability, one can enhance the superconducting transition. This opens up a way to engineer an effective electron-phonon interacting model which suppresses polaronic/charge-density wave tendencies.

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