Editors' Suggestion

Tailoring Superconductivity with Two-Level Systems

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We investigate the impact of two-level systems (TLSs) on superconductivity, treating them as soft modes localized in real space. We show that these defects can either enhance or suppress the superconducting critical temperature, depending on their surface density and average frequency. Using thin-film aluminium as a case study, we quantitatively describe how TLSs modify both the critical temperature and the zero-temperature superconducting gap. Our results thus highlight new opportunities for tailoring material properties through TLS engineering.

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Introduction—The critical temperature T_c of conventional (phonon-exchange) superconductors is principally governed by the electronic band structure and phonon spectrum [1–5]. In materials like aluminium (Al), T_c may be enhanced by introducing structural disorder [6–13] or by approaching the monolayer limit [9,10,14–37]. While the precise origin of this enhancement is debated [38–48], it is generally attributed to one of two mechanisms: (i) modification of the electronic density of states (DoS) or (ii) some change in the electron-phonon spectral function $\alpha^2F(\nu)$. In this Letter, we introduce a third mechanism for enhancing conventional superconductivity: (iii) engineering the density and frequency of two-level systems (TLSs) on the surface of the superconductor. We propose that such a TLS ensemble can be used as a resource for enhancing superconducting coherence.

Two-level systems are localized microscopic defects commonly found in the native oxides of substrate-metal and air-metal interfaces [49–56] and can be viewed as emergent excitations in disordered elastic networks [57–61] (see Fig. 1 for a schematic of TLSs in thin-film aluminium). They serve as a major source of decoherence in superconductor-based quantum technology [55,62–65] and can persist into the epitaxial limit [66–69]. Reasonable agreement between first principles simulations of amorphous Al_2O_3 [70] and experimental studies of polycrystalline γ-Al_2O_3 and amorphous α-AlO_x [71] suggest that localized soft modes (or "vibrons") are a likely candidate for dipole-activated TLSs. As a consequence, a large TLS density will function as an ensemble of dynamical impurities [72,73], with each TLS (vibron) contributing to the low-frequency spectral weight of the bulk phonon population and influencing the superconductor's critical temperature.

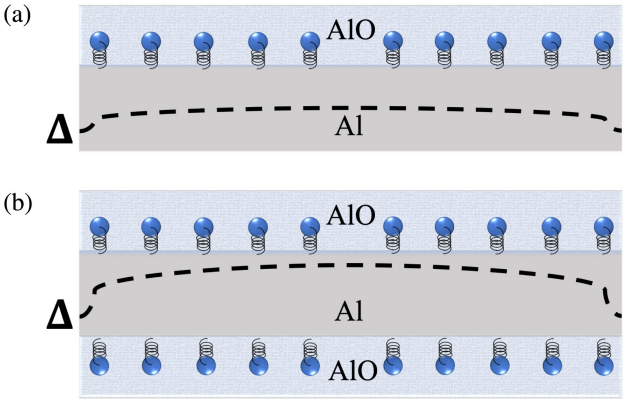


FIG. 1. Two-level systems on (a) the oxidized top surface and (b) the oxidized top and bottom surfaces of an Al sample, raising the local superconducting gap Δ.

We pinpoint under what conditions a finite vibron population will strengthen superconductivity in thin-film Al. This is done by introducing a "two-boson" model where the vibron and bulk phonon contributions to $\alpha^2F(\nu)$ are explicitly taken into account in the calculation of T_c. We focus on multilayer aluminium, with parametrization of the vibron contribution to $\alpha^2F(\nu)$ motivated by I-V data taken on epitaxially grown Al near the monolayer limit [35] and the bulk phonon contribution to $\alpha^2F(\nu)$ found via first

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principles calculations. The critical temperature T_c and the gap edge Δ_1 [74] are then found numerically by solving the Eliashberg equations [46,75–81]. Our work subsequently proposes a new avenue to enhance or suppress superconductivity by engineering the TLS (vibron) frequency [82–85] in such a way as to optimize its contribution to the bulk phonon spectral weight.

Enhancement and suppression of superconducting T_c from two-level systems—Expressions for T_c that take into account the underlying form of the phonon spectrum were put forth by McMillan [2], Allen and Dynes [4], and, most rigorously, Leavens and Carbotte [5,86–88], with the latter deriving $T_c = 1.13\omega_{\ln}\exp[-(1+\lambda)/(\lambda-\mu^*)]$ from the self-consistent Eliashberg equations [5,75,76,89]. Here, μ^* is the Coulomb pseudopotential, λ is the electron-phonon coupling strength, and $\omega_{\ln}$ is the log average phonon frequency [5]. Note that, in general, there is an anticorrelation between λ and $\omega_{\ln}$: namely, that a larger intensity of low-frequency phonons will result in an enhanced λ and a suppressed $\omega_{\ln}$, while a larger intensity of higher-frequency phonons results in the converse [90]. This results in a nontrivial relationship between T_c and the underlying phonon spectrum, which may be further complicated by the varying influence of different phonon frequencies [91].

We model the influence of vibrons on λ, $\omega_{\ln}$, and, ultimately, T_c by adding a new term to the electron-phonon spectral function: namely, $\alpha^2F(\nu) = \alpha^2F^{(0)}(\nu) + n_{\mathrm{TLS}}\alpha^2F_{\mathrm{TLS}}(\nu)$, where n_{TLS} is the density of vibrons (per unit cell per GHz). The full electron-boson coupling λ and log-average boson frequency $\omega_{\ln}$ can be written in terms of the total $\alpha^2F(\nu)$ as

$$\lambda \equiv 2\int_0^\infty d\nu \frac{\alpha^2F(\nu)}{\nu} = \lambda^{(0)} + n_{TLS}\lambda_{TLS} \tag{1a}$$

$$\omega_{\ln} \equiv \omega^{(0)}\exp\left[\frac{2}{\lambda}\int_0^\infty d\nu \frac{\alpha^2F(\nu)}{\nu}\log(\nu/\omega^{(0)})\right], \tag{1b}$$

where $\omega^{(0)}$ is the characteristic phonon frequency of the material in the absence of dynamical defects. In our notation, the (0) superscript always denotes a quantity in the absence of vibrons.

While both $\alpha^2F_{\mathrm{TLS}}(\nu)$ and $\alpha^2F^{(0)}(\nu)$ may have a more complicated structure, a useful first approximation is to assume that both the phonon and vibron contributions to the spectral weight are those of local (Einstein) oscillators; i.e., $\alpha^2F(\nu) = N(0)g^2\delta(\nu-\omega')$, where $N(0)$ is the electronic DoS at the Fermi level, g is the electron-phonon coupling matrix element, and $\omega' = \omega_{\ln} = \omega_E$ (ω_{TLS}) for phonons (vibrons). Such a modification of the bulk electron-phonon spectral function is motivated by our prior work on amorphous Al_2O_3 [70], which provides a first principles motivation for the inclusion of a narrow low-frequency feature to the bulk $\alpha^2F^{(0)}(\nu)$.

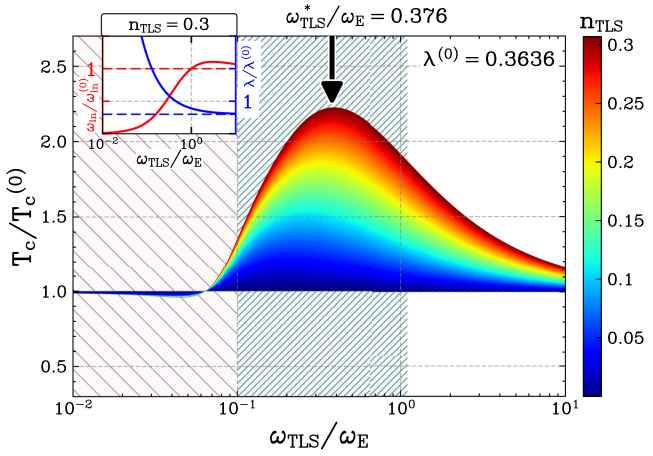


FIG. 2. Change in $T_c/T_c^{(0)}$ versus TLS (vibron) frequency ω_{TLS} normalized by Einstein frequency ω_E and with $\lambda = 0.3636$ held fixed. An Einstein boson model is used for both the bulk phonon and vibron. For $\omega_{\mathrm{TLS}}/\omega_E \geq 0.1$, the critical temperature is found via the Leavens-Carbotte formula [86]. The right-hatched cyan region corresponds to the frequency regime of amplified T_c, with the black arrow marking the maximum T_c, located at $\omega^*_{\mathrm{TLS}}/\omega_E \approx 0.38$. The left-hatched region corresponds to the regime where the Leavens-Carbotte formula is predicted to break down, and thus we suppress T_c to unity for all n_{TLS}. We show numerically in the Supplement that such a breakdown of the Leavens-Carbotte formula is correct [95]. Inset: Comparison of $\omega_{\ln}/\omega_{\ln}^{(0)}$ (red) and $\lambda/\lambda^{(0)}$ (blue) as ω_{TLS} changes for $n_{\mathrm{TLS}} = 0.3$.

A plot of $T_c/T_c^{(0)}$ vs $\omega_{\mathrm{TLS}}/\omega_E$ for $n_{\mathrm{TLS}} = 0.3$ is shown in Fig. 2, with $\omega_{\ln}/\omega_{\ln}^{(0)}$ and $\lambda/\lambda^{(0)}$ plotted in the inset and $\lambda^{(0)} = 0.3636$ (i.e., the bulk electron-phonon coupling we find from first principles). We take the Leavens-Carbotte analytical formula for T_c, as it avoids bulk-specific fitting parameters inherent to the Allen-Dynes theory while still capturing the salient features produced by modifying $\alpha^2F(\nu)$. As μ^* is typically much smaller than $\lambda^{(0)}$, it is ignored for now [88,92,93]. In the limit of $n_{\mathrm{TLS}} \to 0$, we can see from Fig. 2 that $T_c/T_c^{(0)} \to 1$.

For $\omega_{\mathrm{TLS}}/\omega_E < 0.1$, the LC formula predicts that low-frequency vibrons will lower the log average boson frequency, and subsequently result in a severely reduced T_c. This result is unphysical, as it contradicts Anderson's theorem (as a non-magnetic static impurity should have no affect on the critical temperature of an s-wave superconductor [94]). We instead expect vibron modes with $\omega_{\mathrm{TLS}} \ll 2\pi T_c$ to have a reduced influence on the critical temperature, resulting in a breakdown of the Leavens-Carbotte formula [88,91]. For this reason, we interpret the suppression of T_c given by the LC formula to be an artifact of the constant-gap approximation, and should not be interpreted as a genuine physical instability induced by slow TLS modes. In Fig. 2, we artificially suppress the LC formula down to $T_c/T_c^{(0)} \sim 1$ for this low frequency regime (left-hatched), which we'll see later is in agreement

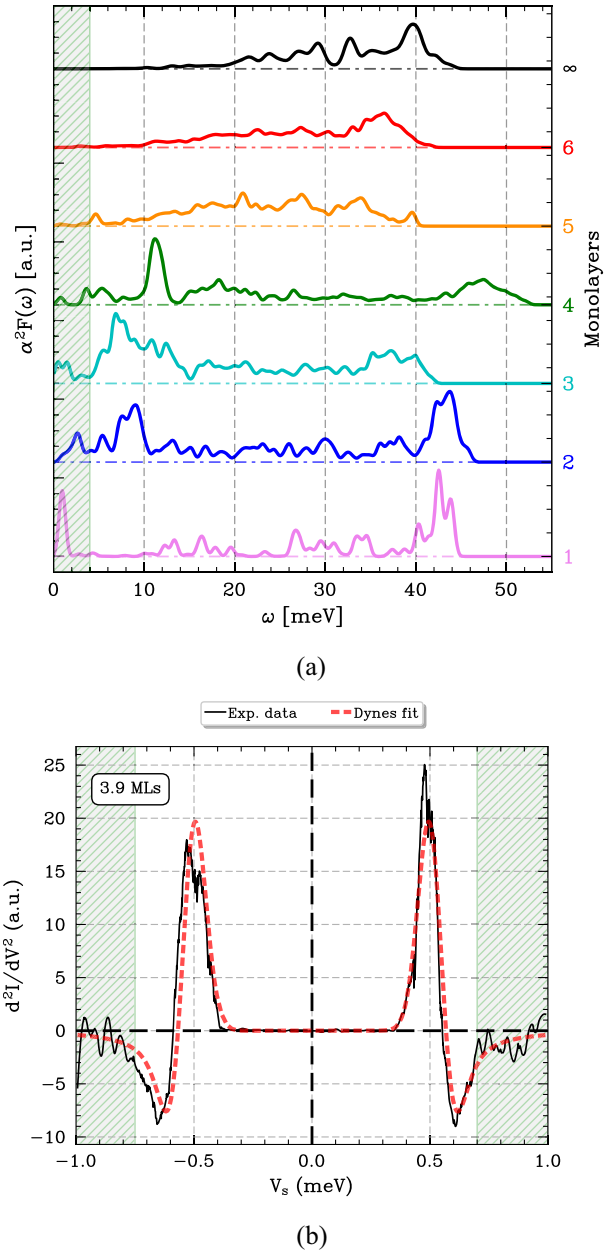


FIG. 3. (a) Electron-phonon spectral function in bulk (top) and multilayered Al found via first principles. In the monolayer limit, more spectral weight is pushed to lower frequencies. The green hatched region corresponds to the regime potentially populated by vibrons ($\omega < 4$ meV). (b) Second derivative of the experimental I-V data taken on an epitaxially grown thin film of Al 3.9 monolayers thick [35] (black) overlaid with the Gaussian-broadened Dynes fit (red-dashed). The voltage regime characterized by localized dips in d^2I/dV^2 is highlighted in green hatch. (a) $\alpha^2F(\nu)$ for bulk and multilayered systems. (b) d^2I/dV^2 for 3.9 monolayer Al.

with numerically solving the linearized Eliashberg equations [95]. For $\omega_{\mathrm{TLS}}/\omega_E > 0.1$, however, note from the inset that both $\omega_{\ln}/\omega_{\ln}^{(0)}$ and $\lambda/\lambda^{(0)}$ remains appreciably large. This results in a "sweet spot" within the window of $0.1 < \omega_{\mathrm{TLS}}/\omega_E < 1$ (right-hatched cyan region of Fig. 2) where the influence of $\alpha^2F_{\mathrm{TLS}}(\nu)$ increases λ but doesn't significantly reduce $\omega_{\ln}$, thereby resulting in an *increase in the superconducting critical temperature*. We will demonstrate that the *vibron-induced enhancement* of superconductivity is not confined to either the constant-gap approximation or the Einstein approximation of the spectral function.

Estimation of T_c from the thin-film electron-phonon spectral function—In order to move beyond the Einstein model and make predictions for more realistic systems, we require more detailed structure for both $\alpha^2F^{(0)}(\nu)$ and $\alpha^2F_{\mathrm{TLS}}(\nu)$. Note that, for the time being, we ignore possible effects from Coulomb interaction or substrate coupling, as we assume the salient consequences of a vibron-enhanced T_c are primarily dependent upon the change of $\alpha^2F^{(0)}(\nu)$ as we approach the thin-film limit.

For $\alpha^2F^{(0)}(\nu)$, we perform first principles simulations on bulk and N-layer Al, with $N = 1$–6. The lattice parameters and atomic positions of all structures are relaxed until all forces are below 1.0×10^{-5} Ry/bohr. Electron-phonon properties are computed on a coarse grid using density functional perturbation theory in the Quantum Espresso software package [96,97] and fully relativistic, norm-conserving pseudopotentials [98,99]. We extract $\alpha^2F^{(0)}(\nu)$ using the EPW package [100–102], interpolating the electron-phonon matrix elements to dense grids in the Wannier basis [103]. Results are shown in Fig. 3(a). Upon decreasing the number of monolayers, a shift in spectral weight toward both the low- and high-frequency regimes is seen, which may be attributed to the simultaneous softening [104,105] and hardening [106] of surface phonons in the single monolayer limit.

Our first principles calculation of $\alpha^2F^{(0)}(\nu)$ does not take into account the presence of vibrons. As such, the addition of the vibron (TLS) contribution to $\alpha^2F^{(0)}(\nu)$ does not result in a double counting of bosonic modes. Note that, although we continue to include the vibron contribution to $\alpha^2F(\nu)$ "by hand," we can justify our more quantitative predictions by turning to experimental data for the estimation of vibron-related parameters. If we assume the vibrons are clustered about a small frequency interval, their net effect on the electron-phonon spectral weight will be a narrow-width, low-frequency Lorentzian [89] addition to $\alpha^2F^{(0)}(\nu)$. The free parameters that go into the determination of $\alpha^2F_{\mathrm{TLS}}(\nu)$ are, thus, the width ϵ_{TLS} of this Lorentzian, the average vibron (TLS) frequency ω_{TLS}, and the TLS density n_{TLS}. Based upon recent experiments [55,65,107–110], n_{TLS} may be estimated to be up to $n_{\mathrm{TLS}} \approx 0.3$ per unit cell within a frequency range of 1 THz (the value used earlier for Fig. 2 [95]). While this is relatively large, it is not unreasonable given experimental data, and allows us to estimate the largest possible modulation of T_c induced by TLSs. A suitable range for TLS frequencies may be inferred by turning to recent STM measurements on

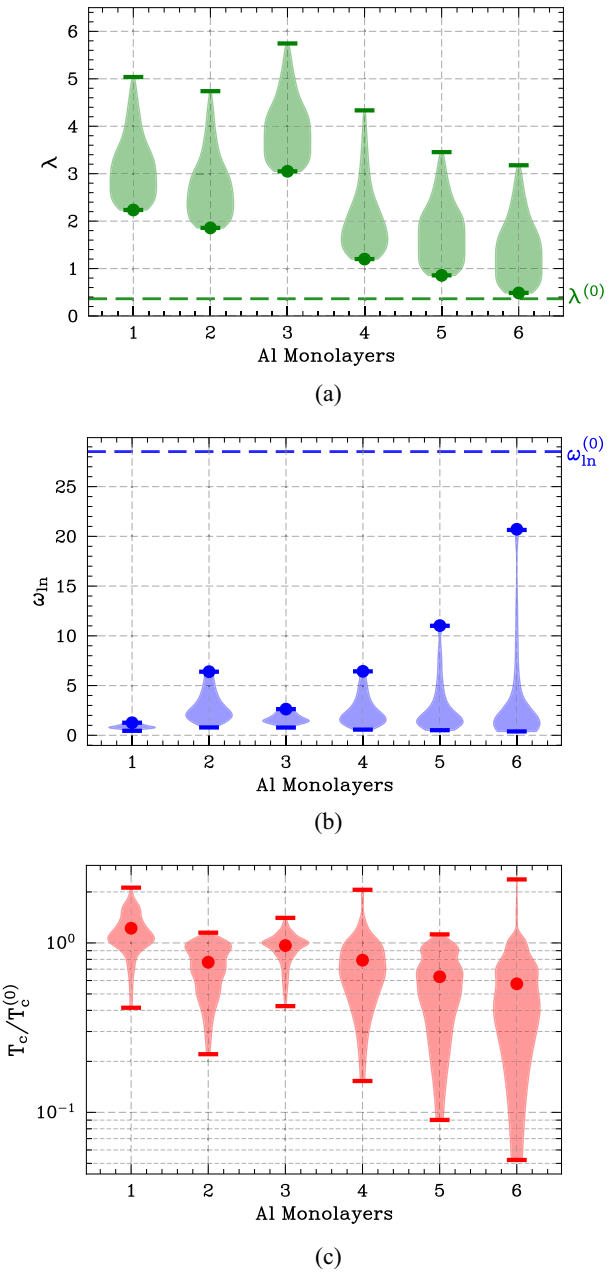


FIG. 4. TLS- or vibron-induced modification of (a) the electron-phonon coupling, (b) the log average phonon frequency, and (c) the ratio of T_c (with vibrons) to $T_c^{(0)}$ (without vibrons) in layered Al. Values for T_c are found via the Leavens-Carbotte equation [86] with $\alpha^2F(\nu)$ found via first principles. The colored error bars correspond to variations of T_c upon changing n_{TLS} and the TLS frequency ω_{TLS}, with the colored shadow representing the spread of data. We use flat top distribution of ω_{TLS} in the range of 0.15 to 4 meV and n_{TLS} in the range 0.01 to 0.3 in generating these plots. As we approach the single monolayer limit, λ grows while $\omega_{\ln}$ shrinks. The net result is larger variation of T_c in the single monolayer limit. As discussed in the main text, the suppression of $T_c/T_c^{(0)}$ in the analytic solution is an artifact of the constant gap approximation taken by Leavens and Carbotte [91]. (a) Electron-phonon coupling λ versus monolayers. (b) Log average frequency $\omega_{\ln}$ versus monolayers. (c) Critical temperature $T_c/T_c^{(0)}$ versus monolayers.

ultrathin epitaxial Al films on Si(111) [35] [see Fig. 3(b)]. As the bias voltage increases to $(\Delta_1 + \hbar\omega_{\mathrm{TLS}})/e$ (where Δ_1 is the gap edge [74]), the resonant exchange of vibrons should enhance the electron-pair binding energy, leading to a local reduction in the effective tunnelling DoS and subsequent dips in d^2I/dV^2 about the TLS (vibron) frequencies [5,111–115]. Such deviations of the measured d^2I/dV^2 [35] from the Gaussian convolution of a Dynes-type fit [116–118] are observed over a broad frequency range extending upward of 0.1 THz above the superconducting gap. While such features in the d^2I/dV^2 profile of thin-film aluminium are not definitive evidence of vibrons, the vibron frequency range suggested from the data in Fig. 3(b) is in agreement with numerical [70] and experimental [119–121] estimates of ω_{TLS} in related materials, thus supporting our underlying hypothesis. We therefore consider values of ω_{TLS} in the frequency range of 0.15–4 meV (around 0.05–1.0 THz) [122]. Upon varying ϵ_{TLS}, we find no large variation in T_c.

The values of λ, $\omega_{\ln}$, and T_c for multilayered Al are shown in Figs. 4(a)–4(c), respectively. For each, we have utilized the Leavens-Carbotte equation with a value of the Coulomb pseudopotential $\mu^* \sim 0.091$ given by first principles [92] and the total $\alpha^2F(\nu)$, including the vibron and bulk contribution, found via our own first principles calculation [123]. As mentioned previously, the severe suppression of T_c predicted by the Leavens-Carbotte formula is an artifact of the constant gap approximation, and is not expected to remain once we numerically solve the linearized Eliashberg equations. Nevertheless, noticeable amplification of the critical temperature (especially in a single monolayer of Al, i.e., aluminene [124]) can be understood as a consequence of phonon softening in the thin film, which pushes significant spectral weight to the GHz range. As suggested in the simple Einstein model, when the phonon frequency is reduced to around $10 \times \omega_{\mathrm{TLS}}$, λ is amplified while $\omega_{\ln}$ remains appreciably large, leading to a boost in T_c.

Finally, note that from the Einstein phonon prediction, we may write the average TLS (vibron) frequency ω^*_{TLS}, which maximizes T_c as $\omega^*_{\mathrm{TLS}} = \alpha\omega_E^{\mathrm{eff}}$. In this equation, $\alpha \lessapprox 0.38$ is a unitless number and a function of n_{TLS} [125], and ω_E^{eff} is an effective Einstein frequency that roughly corresponds to the lowest salient peak(s) in $\alpha^2F(\nu)$. Increasing the TLS frequency $\omega_{\mathrm{TLS}} < \omega^*_{\mathrm{TLS}}$ will result in an increase in T_c, while increasing $\omega_{\mathrm{TLS}} > \omega^*_{\mathrm{TLS}}$ will result in an *apparent* decrease in T_c. While ω_E^{eff} is highly dependent on the precise form of the electron-phonon spectral function, from our *ab initio* calculation [see Fig. 3(a)] and the above equation, we can estimate ω^*_{TLS} around 0.33–1 THz for $n_{\mathrm{TLS}} = 0.3$ and 1–4 monolayers of Al.

Numerical calculation of T_c and the gap edge—Up until this point, we have utilized the Leavens-Carbotte formula for extracting vibron-induced modification of T_c. To more

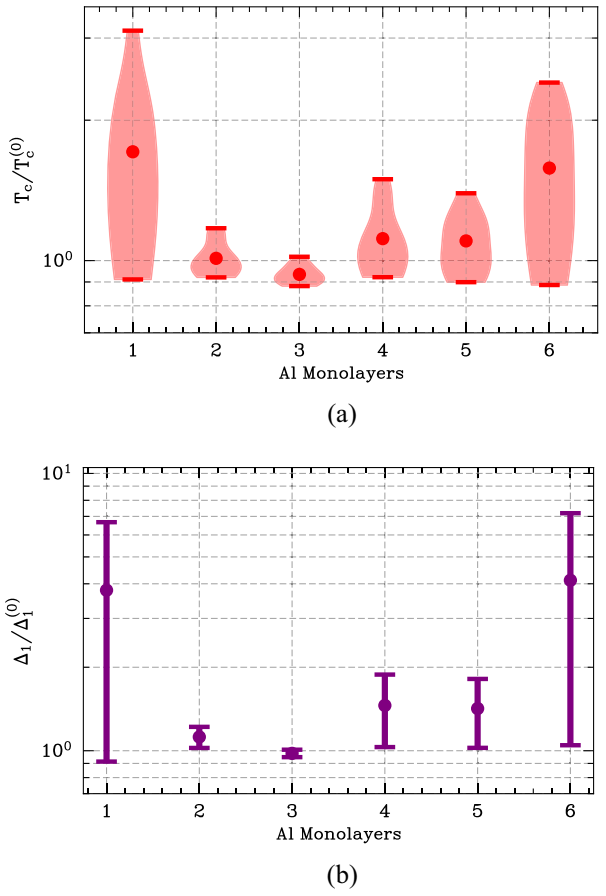


FIG. 5. TLS- and vibron-induced modification of (a) the critical temperature T_c and (b) the gap edge Δ_1. Both T_c and the gap edge are found by numerically solving the Eliashberg equations. Error bars correspond to changes in the respective quantities upon introducing vibrons, and the dots correspond to average values. Vibron parameters are those found in the previous section that led to maximized enhancement and suppression of $T_c/T_c^{(0)}$ when calculated numerically. Compared to the analytical calculation, vibrons tend to enhance (as opposed to suppress) the critical temperature. The change in the local gap edge $\Delta_1/\Delta_1^{(0)}$ follows a similar trend as the critical temperature, except with a more pronounced enhancement. Larger relative enhancement of the gap edge is expected, as the gap-to-T_c ratio is expected to be locally enhanced from the amplified λ induced by the vibrons [129,130]. (a) Critical temperature $T_c/T_c^{(0)}$ versus monolayers, found by numerically solving the Eliashberg equations. (b) Gap edge $\Delta_1/\Delta_1^{(0)}$ versus monolayers, found by numerically solving the Eliashberg equations.

accurately find T_c and the gap, we now turn to numerically solving the Eliashberg equations. The Eliashberg equations may be cast in a local form [90] in which the equation on each real-space "patch" is solved for the local boson frequency or vibron number. In this way, we may talk about local gap enhancement from an inhomogeneous population of vibrons without resorting to the consideration of a full-sample average.

The results of the numerical calculation are shown in Figs. 5(a) and 5(b). In Fig. 5(a), $T_c/T_c^{(0)}$ is found by linearizing the Eliashberg equations on the Matsubara frequency axis and solving the resulting eigenvalue equation via von Mises iteration [5,89,126–128]. TLS and vibron parameters are varied within $\omega_{\mathrm{TLS}} \in [0.1, 4.0]$ meV (slightly lower than experimentally observed), with $n_{\mathrm{TLS}} = 0.3$ held fixed to obtain the maximal effects of a finite vibron population on T_c. We see that the Leavens-Carbotte estimation is most accurate for predicting the maximal $T_c/T_c^{(0)}$ values, with good approximate agreement for $6-2$ Al monolayers. Slight deviations from the analytic prediction of the maximum $T_c/T_c^{(0)}$ may be attributed to the fine structure of $\alpha^2F(\omega)$ [95]. Most notably, however, we observe that the numerical values of $T_c/T_c^{(0)}$ do not exhibit the severe suppression predicted in the analytic solution. As mentioned before, the reduction of $T_c/T_c^{(0)}$ in the presence of low-frequency vibrons is an artifact of the Leavens-Carbotte (and, likewise, the Allen-Dynes) formulation of T_c. As low-frequency vibrons have reduced influence on T_c [91], such deviation from the analytic solution in the presence of low-frequency vibrons is expected and is in agreement with Anderson's theorem [94].

In addition to the critical temperature, we also obtain the $T = 0$ local gap function by numerically solving the Eliashberg equations on the Matsubara frequency axis to obtain $\Delta(i\omega_n)$ and then analytically continuing to real frequencies [131] to get the real axis gap function $\Delta(\omega)$. From the gap function, we can then extract the local gap edge, $\Delta_1 \equiv \Re[\Delta(\omega = \Delta_1)]$ [74]. The smallest TLS frequency is taken to be $\omega_{\mathrm{TLS}} \approx 0.15$, while the maximum TLS frequency is taken to be the value that maximized the value of $T_c/T_c^{(0)}$. Much like the critical temperature, we find that the local gap edge is enhanced by virtue of including TLSs and vibrons; however, in the case of the latter, we find a greater relative enhancement than seen in the former. This is expected, as the ratio of Δ_1 over T_c is expected to range from 1.76 in the BCS limit [132] to ~6–6.5 in the very strongly coupled limit [5,129]. As such, an increase of $\Delta_1/\Delta_1^{(0)} \sim 7$ is not unreasonable if we assume $T_c/T_c^{(0)} \sim 2.5$–3.

Conclusions—By treating two-level systems as localized soft modes (i.e., vibrons), we have shown that TLSs add to the spectral weight of bulk phonons, directly influencing the critical temperature and gap of the superconductor. As quantum confinement effects alone cannot explain the large boost in T_c observed in epitaxial Al films grown on Si(111) [35,81], extreme enhancement of the critical temperature as a function of film thickness may be a result of dimensional reduction in tandem with a finite vibron population. As a whole, we emphasize that our work opens new avenues for TLS-based material engineering and phase stabilization, and consequently, shifts the focus of TLSs from a ubiquitous source of decoherence to an abundant resource for enhancing superconductivity in the two-dimensional limit.

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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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