Limitations on Electronic Mechanisms for High Temperature Superconducting Oxides

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ABSTRACT

The effects of electronic mechanisms for electron pairing in high temperature superconducting oxides on both the transition temperature T_c and the isotope effect parameter α are considered. It is shown that for the higher T_c oxides, measured values of T_c and α together with estimates of the phonon contributions are not consistent with high-frequency electronic mechanisms. Limitations of the theory and some constraints on low-frequency electronic mechanisms are also discussed.

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Even before the discovery of high temperature superconductivity (HTS) in the oxides,[1] there were many theoretical suggestions for raising the superconducting transition temperature T_c. Most of the theoretical suggestions were based on BCS theory[2] and focused on mechanisms using pairing bosons other than phonons. Electronic mechanisms were favored primarily because their high frequency nature leads to interaction energy cutoffs which are much larger than phonon energies. Since the energy cutoffs become prefactors in standard equations for T_c, these mechanisms yield very high estimates of T_c. When the role of the energy cutoffs is included in the pairing parameters, it can be shown[3] that having the highest possible frequencies may not maximize T_c. Nevertheless in general[4] higher frequency mechanisms lead to higher T_c's if no other restrictions are imposed.

The specific nature of the electronic mechanism depends on the models used. Examples include collective excitations based on exciting electron-hole pairs, excitons, plasmons, acoustic plasmons or demons, spin fluctuations, and charge transfer excitations which are all electronic in nature and can give rise to electron pairing. These mechanisms were among the first to be suggested[5-7] for explaining the high T_c's observed in the HTS oxides. More recently the number of suggestions of mechanisms of this kind has increased substantially.

At present there is no consensus on the dominant mechanism causing HTS in the oxides. The purpose of the present study is to use measured data and theoretical estimates to constrain the theories and limit the possibilites. An

earlier attempt[8] in this direction emphasized the observation that because high frequency mechanisms are only consistent with weak coupling BCS theory, experimental tests of the coupling strength can be used to determine the viability of these mechanisms. Although measurements such as the ratio of the energy gap to T_c have been made using IR spectroscopy, Raman scattering, tunneling, spin resonance, and photoemission experiments, no definitive conclusion has been reached. Hopefully the scatter in these results will be reconciled with a consistent model in the future.

The approach here is to examine the limitations placed on the theory by current data. To do this, several models are used to calculate T_c and the isotope effect parameter $\alpha = -(d\log T_c)/(d\log M)$. For each case, the kernel of the attractive interaction contains a model electronic interaction as well as a BCS-like electron-phonon contribution. The values of the phonon and electronic coupling strengths are found to be limited when comparison is made with experimental data for T_c and α. These limits can then be used to constrain the theories.

Within a restricted range of coupling strength λ, we can employ analytic solutions of approximate models to try to constrain theories employing an electronic mechanism. For example, in the weak coupling limit $\lambda \lesssim 1$, we consider a McMillan[4] equation with a prefactor ω_{log} where[4,9]

$$\omega_{log} = \lim_{n\to 0}\langle\omega^n\rangle^{1/n} = \exp\langle\ln\omega\rangle \qquad (1)$$

and a δ-function model of $\alpha^2 F(\omega)$

$$2\alpha^2 F(\omega) = \sum_{i=1}^{m} \lambda_i \omega_i \delta(\omega - \omega_i) + \lambda_e \omega_e \delta(\omega - \omega_e) \qquad (2)$$

where m is the number of phonon peaks of strength λ_i and frequency ω_i, and λ_e and ω_e represent the strength and frequency of the electronic peak. A calculation of the isotope effect parameter α with the Coulomb repulsion $\mu^* = 0$ yields

$$\alpha = \frac{\lambda_p}{2\lambda} \qquad \text{where} \quad \lambda_p = \sum_{i=1}^{m} \lambda_i. \qquad (3)$$

Taking account of the Coulomb repulsion will tend to decrease α.

A three-square-well model[10,11] can be used in the very weak coupling case, for which we find that λ_p/λ is approximately constant for $\omega_{el} \gg T_c$ when ω_{el} is varied while holding T_c and α constant. In the simple case where $\mu^* = 0$,

$$\frac{\lambda_p}{\lambda} = \left(1 + \frac{C_1}{C_2 + \ln(\omega_{el}/\omega_{ph})}\right)^{-1} \qquad (4)$$

where C_1, C_2 are constants of order unity and are determined by the choice of T_c and α. Thus, λ_p/λ varies slowly both in the weak coupling and intermediate coupling range for $\mu^* = 0$. Numerical solutions of the three-square-well model for $\mu^* \neq 0$ show the same behavior of λ_p/λ for $\omega_{el} \gg T_c$.

From Eqs. (3) and (4), it is clear that the coupling strength ratio λ_p/λ is constrained by T_c and α, particularly for the McMillan equation with the ω_{log} prefactor for which the ratio is independent of ω_{el}. We therefore consider some of the experimental data available for the HTS oxides to see if the data

are consistent with these theoretical constraints. Since a wide range of λ is required, the Eliashberg[12] equations are solved together with models of the interaction kernels to determine T_c and the isotope effect parameter α. This method yields the most reliable state-of-the-art solutions over the entire range of λ considered. The input required is the standard spectral function $\alpha^2F(\omega)$ which normally is interpreted as an electron-phonon matrix element weighted by the phonon density of states $F(\omega)$. Here we assume that both the phonon and electronic pairing mechanisms can be described in this way with the caveat that Migdal's theorem[4] is assumed for both but may not be valid for the electronic contribution. The Coulomb repulsion is modeled in the standard way with the parameter μ^* which is forced to be 0.13 throughout the calculation.

The Eliashberg equations are solved in the Matsubara representation[4,9,13] which requires diagonalization of matricies with sizes determined by the ratio of the highest energy cutoff of $\alpha^2F(\omega)$ divided by T_c. Similar calculational approaches[14-16] were used earlier to model the interactions in the oxides; however the use of a negative μ^* for modeling an attractive electronic mechanism as done in reference 14 is not done here.

Both the phonon and electronic features in $\alpha^2F(\omega)$ are modeled with Lorentzian peaks of varying strength. The phonon contribution is evaluated by choosing a few peaks to mimic the estimates from experiments on the oxides. The electronic contribution is represented by a single peak with variable height

and energy. An electron-phonon coupling parameter λ_p and an electronic coupling parameter λ_e are obtained by integrating over $\alpha^2F(\omega)$ in the appropriate region $\Delta\omega$ using

$$\lambda_{\Delta\omega} = 2\int_{\Delta\omega} \alpha^2F(\omega)\frac{d\omega}{\omega}. \tag{5}$$

By choosing $\Delta\omega$ appropriately the coupling parameters λ_p, λ_e, and $\lambda = \lambda_p + \lambda_e$ are evaluated for a given $\alpha^2F(\omega)$. The isotope effect parameter α is determined by assigning an ionic mass dependence to the various parts of the phonon spectrum. In this way the entire phonon spectrum or parts can be made to shift with changes in the isotopic mass.

Three generic cases are considered, corresponding to (I) the Y–Ba–Cu–O system[17,18] with $T_c \sim 93$K and $\alpha = 0.02$, (II) the La–(Ba,Sr)–Cu–O system[19,20] with $T_c \sim 40$K and $\alpha = 0.15$ and (III) the Ba–K–Bi–O system[21] with $T_c \sim 30$K and $\alpha = 0.41$. In each case T_c and α are held constant and assumed to be given by experiment and the appropriate λ_e and λ_p are evaluated as a function of the energy of the electronic peak for a given phonon spectrum.

Some of the results are contrary to what is expected for these systems. For example, the calculations reveal that a high frequency electronic mechanism can be added to the phonon induced pairing interaction for the lowest T_c system (Case III), but it is contrary to the data for the highest T_c system (Case I). For Case III, our calculations indicate that the experimental data do not restrict the choice of an electronic mechanism. We therefore focus on the higher T_c

oxide superconductors. It is usually assumed that Cases I and II require an added high frequency electronic mechanism to achieve the high T_c, but this is inconsistent with the results for α and estimates of λ_p as shown below.

For case I, the phonon peaks for $\alpha^2F(\omega)$ were chosen to represent the measured $F(\omega)$ spectrum.[22] It was found that this could be done with two peaks at 17 and 70 meV with weights of 1 and 2 respectively. The solution of the Eliashberg equations is shown in Fig. 1 where λ_p and λ_p/λ are displayed as a function of the energy position of the electronic peak. The weight of the electronic peak and λ were adjusted at each frequency to yield the measured values of T_c and α. The most dramatic result is the near constancy of λ_p/λ above 100 meV. Hence, for fixed T_c and α the ratio λ_p/λ is nearly independent of the position of the electronic peak at high energy, and the constraints on T_c and α forces λ_p to be small, $0.05 \leq \lambda_p \leq 0.10$. Estimates[23] of λ_p range from a factor of 3 to an order of magnitude larger than the upper limit found here. This implies that for the model assumed here, an electronic mechanism with a characteristic energy above 100 meV is inconsistent with the measurements of T_c, α, and the estimates of λ_p. A low-energy electronic mechanism can also be excluded since $0.1 < \lambda_p < 0.3$ for ω_{el} between 10 and 100 meV. Therefore the expected presence of a phonon pairing mechanism together with conservative estimates of a minimum electron-phonon coupling rule out an electronic pairing mechanism for the model used to describe Case I.

Similar but less restrictive results are found for Case II. The $\alpha^2F(\omega)$ spectrum is again based on experimental data[24] and is modeled by Lorentzian peaks at 25, 45, and 80 meV with strengths 2, 1.5, and 1 respectively. The results are given in Fig. 2. Again the ratio λ_p/λ is fairly constant for frequencies > 70 meV, but λ_p can be larger ($0.15 \leq \lambda_p \leq 0.30$) in this range. As in Case I, this range of λ_p is a factor of 2 to an order of magnitude lower than estimated.[25] This implies that a high energy electronic peak is also inconsistent with the data for Case II superconductors.

The results for Cases I, II, and III suggest that the data for materials in these three classes are relevant to the appropriateness of a high frequency electronic pairing mechanism. Specifically, a high frequency pairing mechanism independent of ionic mass can be added to the phonon induced pairing interaction for the low T_c materials, but this is not feasible for Cases I and II. This is opposite to the widely held view that if high frequency electronic mechanisms are involved, they are more likely for Case I than for Case III materials.

Estimates of λ and λ_p vary widely in the HTS. Experimentally, small values of λ have been derived from the linearity of the resistivity $\rho(T)$ over a large temperature range. We do not use these estimates for constraints within our model because a clear separation between the electronic and phonon contributions is not possible. However, it is likely that the linearity of $\rho(T)$ argues against the existence of a high-energy electronic mechanism, since such a mechanism should also cause nonlinearity in $\rho(T)$ up to temperatures of order

$\hbar\omega_{el}/k_B$. Theoretical estimates of λ in La–(Ba,Sr)–Cu–O[25] range from 2.5 to 3.8, while the estimates for Y–Ba–Cu–O[23] range from 0.32 to 1.3. Both these ranges are far above the range of λ_p found above, excluding the possibility that a high-energy electronic mechanism can explain HTS within our model for Cases I and II.

It is important to emphasize that we have calculated the transition temperature T_c and the isotope effect α using isotropic, three-dimensional Eliashberg theory with a model electron-phonon interaction spectrum For the model spectrum, sets of two or three peaks were chosen to mimic $\alpha^2F(\omega)$ or $F(\omega)$ from experiment, and a high energy peak independent of ionic mass was added to the spectrum to simulate the addition of an electronic pairing mechanism. We have also performed similar calculations using a numerical $F(\omega)$ derived from force constant fitting to the experimental phonon density of states for La_2CuO_4. These calculations showed the same behavior of λ_p/λ, indicating our results are not an artifact of the simplified nature of our $\alpha^2F(\omega)$.

In Cases I and II, we are unable to obtain the experimental values for T_c and α unless the electron-phonon coupling λ_p is much weaker than theoretically predicted. For Y–Ba–Cu–O and La–(Ba,Sr)–Cu–O, therefore, within the approximations, our calculations eliminate the possibility that a high energy electronic mechanism can explain HTS within an isotropic three-dimensional model. In Y–Ba–Cu–O, low-energy electronic mechanisms can also be excluded.

For the Ba–K–Bi–O superconductor, we are unable to exclude an electronic mechanism at any energy scale based on our calculations.

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FIGURE CAPTIONS

FIG. 1. Calculated values of λ_p/λ (squares) and λ (triangles) for Y–Ba–Cu–O plotted against the energy of the electronic mechanism.

FIG. 2. Calculated values of λ_p/λ (squares) and λ (triangles) for La–(Ba,Sr)–Cu–O plotted against the energy of the electronic mechanism.

Fig. 1

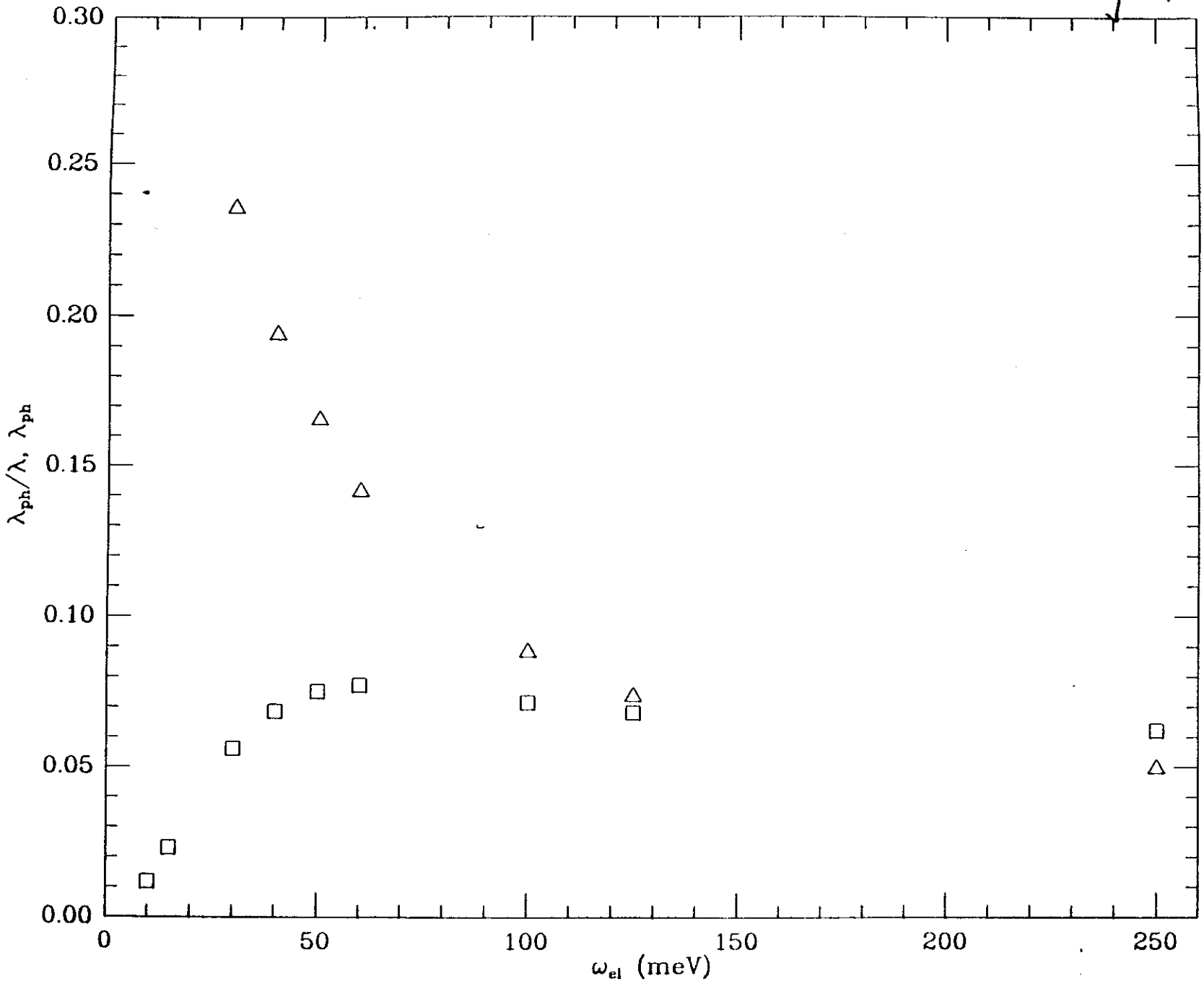
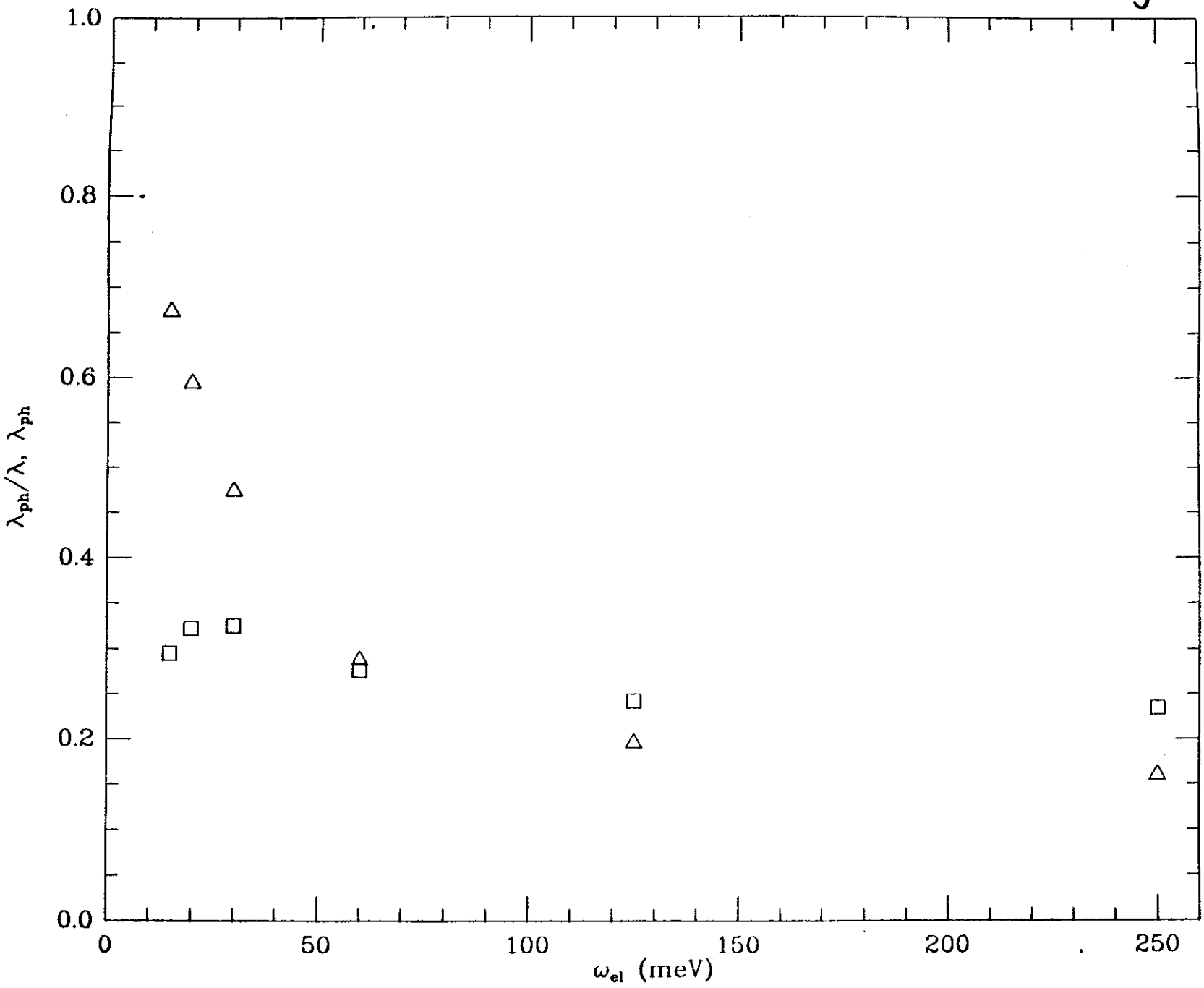

0.30
0.25
0.20
0.15
0.10
0.05
0.00
λ_{ph}/λ, λ_{ph}
0
50
100
150
200
250
ω_{el} (meV)

Fig. 2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0.8
0.6
0.4
0.2
0.0
0
50
100
150
200
250
λ_{ph}/λ, λ_{ph}
ω_{el} (meV)