Upper bound on the specific-heat jump: Application to the high-T_c oxides

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We establish an upper limit on the normalized specific-heat jump $\Delta C/\gamma(0)T_c$ at T_c, for the case of an isotropic Eliashberg superconductor. Functional derivative results for $\Delta C/\gamma(0)T_c$, in the case of realistic electron-phonon spectral densities, lead us to consider a model delta-function spectrum which, we can prove, gives a local maximum in $\Delta C/\gamma(0)T_c$ for some low but finite phonon energy. The value of the maximum depends somewhat on the value of Coulomb pseudopotential (μ^*) used, but is always surprisingly small. As an example for $\mu^*=0.15$, $\Delta C/\gamma(0)T_c \leq 3.73$ with the inequality holding for realistic spectra. This is smaller than some recent experimental values published for $La_{1.85}Sr_{0.15}CuO_4$.

I. INTRODUCTION

The recent discovery of superconductivity in La-Ba-Cu-O by Bednorz and Müller[1] has led to great interest in related systems and to the discovery of superconductivity above 90 K in Y-Ba-Cu-O.[2] A great deal of experimental information is already available in the published literature[3] as well as in unpublished form. It is well established that the mechanism responsible for superconductivity, in most conventional metallic systems, is the electron-phonon interaction. While some evidence exists that, at least for $La_{1.85}Sr_{0.15}CuO_4$,[4-6] the mechanism may be the same, it is by no means, as yet, completely certain, and rival theories[7-10] have been put forward. Thus, the question of the nature of the mechanism involved, which is fundamental, remains open.

In this paper we pose and attempt to answer, partially, the following question. For an electron-phonon superconductor, described by the Eliashberg equations,[11-14] with arbitrary strength of the electron-phonon spectral density $\alpha^2F(\omega)$, is there a maximum value for the electronic specific-heat jump $\Delta C(T_c)$ at T_c normalized by the "normal-state specific heat" $\gamma(0)T_c$ [where $\gamma(0)$ is Sommerfeld constant]? The answer to this question could help rule out or confirm the electron-phonon interaction as the mechanism in a particular case. For example, if $\Delta C(T_c)/\gamma(0)T_c$ is firmly established, experimentally, as larger than any maximum that may exist for an Eliashberg superconductor, this could be taken as strong evidence against such a mechanism. It should be stressed that in the approach taken in this paper, no attempt is made to understand whether or not the spectral densities used are consistent with lattice stability or, for that matter, with Migdal's theorem. We simply proceed as if this were the case. The free energy is discussed in Sec. II and functional derivatives are introduced in Sec. III. A scaling theorem is formulated and proved in Sec. IV, followed by results (Sec. V), and conclusions in Sec. VI.

II. FREE ENERGY

The free-energy difference between the normal and superconducting states in Eliashberg theory is given by the formula[11-14]

$$\Delta F = 2\pi N(0)k_BT\sum_{n>0}\left[2\left(\left[\tilde{\Delta}^2(i\omega_n)+\tilde{\omega}^2(i\omega_n)\right]^{1/2}-\tilde{\omega}(i\omega_n)-\frac{1}{2}\frac{\tilde{\Delta}^2(i\omega_n)}{\left[\tilde{\Delta}^2(i\omega_n)+\tilde{\omega}^2(i\omega_n)\right]^{1/2}}\right)-\left[\tilde{\omega}(i\omega_n)-\tilde{\omega}^0(i\omega_n)\right]\left(\frac{\tilde{\omega}(i\omega_n)}{\left[\tilde{\Delta}^2(i\omega_n)+\tilde{\omega}^2(i\omega_n)\right]^{1/2}}-1\right)\right], \tag{1}$$

where $N(0)$ is the single spin density of electron states at the Fermi surface and k_B is the Boltzmann constant. The $\tilde{\Delta}(i\omega_n)$ and $\tilde{\omega}(i\omega_n)$ are, respectively, the Matsubara pairing energy function and renormalized frequencies with $i\omega_n \equiv i\pi T(2n-1)$, $n=0, \pm 1, \pm 2, \ldots$, and T is the temperature. The superscript zero on one $\tilde{\omega}(i\omega_n)$ in Eq. (1), denotes that the normal-state value of this quantity is to be taken.

The two coupled nonlinear Eliashberg equations on the imaginary frequency axis are well known[11-14] and have the form

$$\tilde{\Delta}(i\omega_n)=\pi T\sum_m\left[\lambda(m-n)-\mu^*\Theta(\omega_c-|\omega_m|)\right]\times\frac{\tilde{\Delta}(i\omega_m)}{\left[\tilde{\Delta}^2(i\omega_m)+\tilde{\omega}^2(i\omega_m)\right]^{1/2}}, \tag{2}$$

and

$$\tilde{\omega}(i\omega_n)=\omega_n+\pi T\sum_m\left[\lambda(m-n)\right]\frac{\tilde{\omega}(i\omega_m)}{\left[\tilde{\Delta}^2(i\omega_m)+\tilde{\omega}^2(i\omega_m)\right]^{1/2}}, \tag{3}$$

with

$$\lambda(m-n)=2\int_0^\infty \frac{\omega\alpha^2F(\omega)\,d\omega}{\omega^2+(\omega_n-\omega_m)^2}\ . \tag{4}$$

It is clear that Eqs. (2) and (3) depend only on the electron-phonon spectral density. This is because the Coulomb repulsion pseudopotential μ^* in our work is fixed at several convenient values and the linearized version of the Eliashberg equations, in the Matsubara representation, then gives the critical temperature T_c. The cutoff in μ^* is not an extra parameter but is fixed at some large value. In principle, some choice of electronic density of states $N(0)$ is also needed if one wants the free energy, but this does not enter if we are interested only in a normalized ratio, such as the specific-heat jump $\Delta C(T)$ at T_c divided by the "normal-state specific heat" $\gamma(0)T_c$. It should perhaps be stressed, at this point, that $\gamma(0)T$ is not, strictly speaking, the "normal-state electronic specific heat at temperature T" because, in general, $\gamma(T)$ is temperature dependent when the electron-phonon interaction[15-17] is taken into account. Thus, $\gamma(0)T$ is the zero-temperature Sommerfeld constant multiplied by T. While, for conventional superconductors, $\gamma(0)$ and $\gamma(T_c)$ are not very different, when T_c gets very high this is no longer the case.[5] Still, we will follow convention and use $\gamma(0)T_c$ for normalization purposes. This is attractive because it is $\gamma(0)$ and not $\gamma(T_c)$ that comes into the slope of the upper critical field at T_c, a quantity that is often used to extract an estimate of the Sommerfeld constant. The normal-state electronic specific heat itself at T_c would be extremely difficult to measure, even in principle, since it is superimposed on, and strongly coupled to, a large unknown phonon background in high-T_c superconductors.

III. FUNCTIONAL DERIVATIVES

To understand how we might proceed from here, we consider first the functional derivative with electron-phonon spectral density $\alpha^2F(\omega)$ of $\Delta C(T_c)/\gamma(0)T_c$. Marsiglio, Schachinger, and Carbotte[18] give the formula

$$\frac{1}{\gamma(0)}\frac{\delta[\Delta C(T)/T_c]}{\delta\alpha^2F(\omega)}=\frac{N(0)}{\gamma(0)}\frac{T}{T_c}\frac{d^2}{dT^2}\left[(\pi T)^2\sum_{n,m}\left(\frac{\tilde\omega(n)}{[\tilde\omega^2(n)+\tilde\Delta^2(n)]^{1/2}}\frac{\tilde\omega(m)}{[\tilde\omega^2(m)+\tilde\Delta^2(m)]^{1/2}}-\mathrm{sgn}(\omega_n\omega_m)\right.\right.$$
$$\left.+\frac{\tilde\Delta(n)}{[\tilde\omega^2(n)+\tilde\Delta^2(n)]^{1/2}}\frac{\tilde\Delta(m)}{[\tilde\omega^2(m)+\tilde\Delta^2(m)]^{1/2}}\right)\times\left(\frac{2\omega}{\omega^2+(\omega_n-\omega_m)^2}-\frac{2}{T_c}(\omega_n-\omega_m)^2\frac{\delta T_c}{\delta\alpha^2F(\omega)}\int\frac{2\,d\omega'\omega'\alpha^2F(\omega')}{[\omega'^2+(\omega_n-\omega_m)^2]^2}\right)\Bigg]\ , \tag{5}$$

from which

$$\frac{\delta}{\delta\alpha^2F(\omega)}\left(\frac{\Delta C(T_c)}{\gamma(0)T_c}\right)$$

follows using the relation

$$\frac{1}{\gamma(0)}\frac{\delta}{\delta\alpha^2F(\omega)}\left(\frac{\Delta C(T_c)}{T_c}\right)-\frac{1}{\gamma(0)}\frac{\delta\gamma(0)}{\delta\alpha^2F(\omega)}\left(\frac{\Delta C(T_c)}{\gamma(0)T_c}\right)\ , \tag{6}$$

where

$$\delta\gamma(0)/\delta\alpha^2F(\omega)=\tfrac{2}{3}\pi^2k_B^2N(0)2/\omega\ .$$

This last result follows from the usual formula $\gamma(0)=\frac{2}{3}N(0)\pi^2k_B^2(1+\lambda)$ with λ the electron-phonon mass renormalization given by formula (4) with $n=m$.

As an example, functional-derivative results for the case of Pb are presented in Fig. 1 (solid curve). What is plotted is $(1+\lambda)T_c$, multiplied by the functional derivative of interest, as a function of phonon energy ω normalized to the critical temperature T_c (k_B—the Boltzmann constant sometimes taken to be one in this work). The divergence toward $-\infty$, at low ω, can be traced to the $1/\omega$ factor in the second term of Eq. (6) and comes directly from our use of $\gamma(0)$ as normalization factor. If instead $\gamma(T_c)$ were used, as in the work of Marsiglio *et al.*,[18] the divergence would not be present and the corresponding functional derivative would go smoothly to zero as $\omega\to 0$. At higher frequencies, around $\omega/T_c\cong 5$, a positive maximum occurs and then the functional derivative slowly decays towards zero as $\omega\to\infty$. We have tried many other realistic tunneling-derived spectra[14] instead of Pb, and find, in all cases, the same shape for the functional derivative.[18] For example, the dotted line was calculated using the Al spectrum given by Leung, Carbotte, Taylor, and Leavens.[19] It is very close to the dashed line which was calculated using a two-square-well model for $\lambda(n-m)$. Details of the two-square-well model results can be found in the work of Marsiglio and Carbotte.[20] In this simplified model, no assumption is made about the shape of the spectral density, except that all important phonon frequencies should be much greater than several k_BT_c's. This is the usual BCS limit, and applies to all weak coupling systems.

From the above, we conclude that the shape of

$$\delta[\Delta C(T_c)/\gamma(0)T_c]/\delta\alpha^2F(\omega)$$

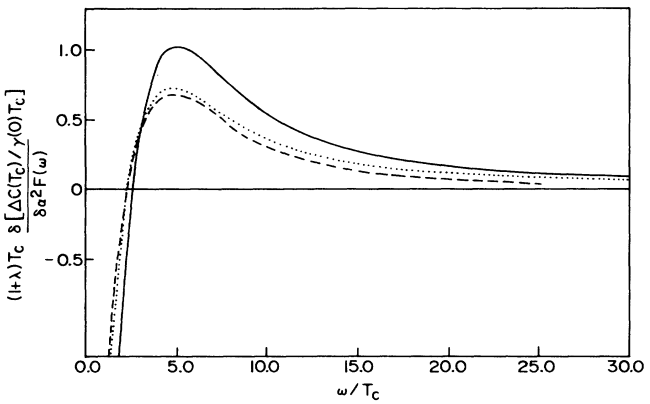

FIG. 1. The functional derivative of the specific-heat jump at T_c normalized to $\gamma(0)T_c$, where $\gamma(0)$ is the Sommerfeld constant. What is given is $(1+\lambda)T_c\delta[\Delta C(T_c)/\gamma(0)T_c]\delta\alpha^2F(\omega)$ vs ω/T_c. The solid curve is for Pb, the dotted line for Al, and the dashed line for any weak coupling superconductor in a two-square-well model.

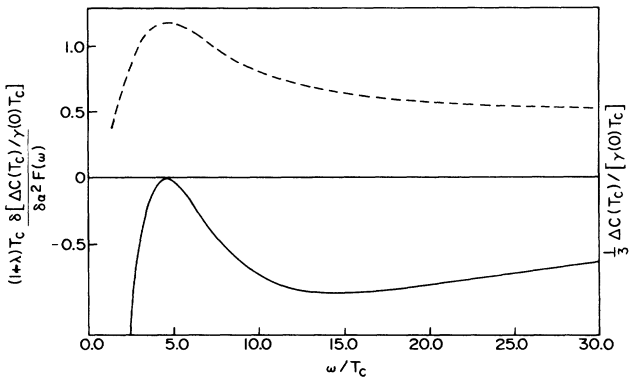

FIG. 2. The value of $\frac{1}{3}\Delta C(T_c)/[\gamma(0)T_c]$ (right-hand label) for an Einstein spectrum as a function of Ω_E/T_c (dashed curve), where Ω_E is the position of the phonon frequency. The results are independent of the value used for the area (A) under the delta function. The μ^* value was 0.051. The functional derivative $\delta[\Delta C(T_c)/\langle\gamma(0)T_c\rangle]/\delta\alpha^2F(\omega)$ multiplied by $(1+\lambda)T_c$ (left-hand label) for the case of a delta-function spectrum $\alpha^2F(\omega)=A\delta(\omega-\Omega_E^*)$, where Ω_E^* is the frequency of the maximum in the dashed curve. Note that it is negative definite and exactly zero at the frequency Ω_E^* of the optimum spectra. This indicates a local maximum.

is fairly universal for realistic shapes of $\alpha^2F(\omega)$ (i.e., actual measured shapes for real materials[12,14]). This leads to the suggestion that in order to increase $\Delta C(T_c)/\gamma(0)T_c$ for a given $\alpha^2F(\omega)$, we should take weight from some frequency where the functional derivative is smaller than its value at maximum, and transfer it to the optimum frequency, keeping the total area under $\alpha^2F(\omega)$ constant. This suggests that to maximize $\Delta C(T_c)/\gamma(0)T_c$, for a given value of $A=\int\alpha^2F(\omega)\,d\omega$, we should use a delta function with all its weight placed at the same Einstein frequency Ω_E.

In Fig. 2 we show results of such calculations for $\mu^*=0.051$. What is plotted (dashed curve) is the dimensionless ratio $\frac{1}{3}\Delta C(T_c)/\gamma(0)T_c$ as a function of Ω_E/T_c (right-hand side label). It is seen that, as we expected from our functional derivative arguments, on lowering the position of the delta function Ω_E/T_c from high frequencies, the value of $\Delta C(T_c)/\gamma(0)T_c$ increases until a maximum of 3.57 is reached for $\Omega_E^*=4.55T_c$. Note that, after the maximum, the ratio drops rather fast towards very small values as ω goes to zero. Note also, before moving on, that the value of $\Delta C(T_c)/\gamma(0)T_c$ at maximum is not very much larger than the value observed in some real systems.[14] We will return to this fact later.

IV. A SCALING THEOREM

At this point, it is necessary to stress that our estimate for $\Delta C(T_c)/\gamma(0)T_c$ at maximum is independent of the value chosen for A—the area under the delta-function model spectrum, except for small corrections due to a finite μ^*. This follows from a scaling law, originally discussed by Leavens,[21] for T_c. This work was extended to include paramagnetic impurities by Ashraf and Carbotte,[22] and to below T_c by Carbotte, Marsiglio, and Mitrović.[23] For an $\alpha^2F(\omega)=A\delta(\omega-\Omega_E)$, the Eliashberg equations (2) and (3) can be written in the dimensionless form

$$\tilde{\bar{\Delta}}(i\omega_n)=\pi k_B\bar{T}\sum_m\left\{\frac{2\bar{\Omega}_E}{\bar{\Omega}_E^2+(\bar{\omega}_n-\bar{\omega}_m)^2}-\mu^*\Theta(\omega_c-|\bar{\omega}_m|A)\right\}\frac{\tilde{\bar{\Delta}}(i\omega_m)}{[\tilde{\bar{\Delta}}^2(i\omega_m)+\tilde{\bar{\omega}}^2(i\omega_m)]^{1/2}} \tag{7}$$

and

$$\tilde{\bar{\omega}}(i\omega_n)=\bar{\omega}_n+\pi k_B\bar{T}\sum_m\frac{2\bar{\Omega}_E}{\bar{\Omega}_E^2+(\bar{\omega}_n-\bar{\omega}_m)^2}\frac{\tilde{\bar{\omega}}(i\omega_m)}{[\tilde{\bar{\Delta}}^2(i\omega_m)+\tilde{\bar{\omega}}^2(i\omega_m)]^{1/2}}\ , \tag{8}$$

with $\bar{T}\equiv T/A$, $\bar{\Omega}_E\equiv\Omega_E/A$, $\tilde{\bar{\omega}}(i\omega_m)\equiv\tilde{\omega}(i\omega_m)/A$, and $\tilde{\bar{\Delta}}(i\omega_m)\equiv\tilde{\Delta}(i\omega_m)/A$. It is clear from (7) and (8) that $\tilde{\bar{\Delta}}$ and $\tilde{\bar{\omega}}$ are independent of A, provided we neglect a very small correction from the cutoff ω_c in the Coulomb repulsion μ^*, as done by Leavens.[21] Also, referring to formula (1) for the free energy, we see that ΔF is proportional to $N(0)$ and to A^2 with the proportionality factor a function of $\bar{\Omega}_E$ and $\bar{T}$ only. Thus, $\Delta F=N(0)A^2f(\bar{\Omega}_E,\bar{T})$, where the function f can be identified from Eqs. (1), (7), and (8). Thus, the specific-heat difference

$$\Delta C(T)\equiv T\frac{d^2\Delta F}{dT^2}=\bar{T}AN(0)\frac{d^2f(\bar{\Omega}_E,\bar{T})}{d\bar{T}^2}\ , \tag{9}$$

and therefore

$$\frac{\Delta C(T_c)}{\gamma(0)T_c}=\frac{3}{2\pi^2 k_B^2(1+2/\bar{\Omega}_E)}\frac{d^2 f(\bar{\Omega}_E,\bar{T})}{d\bar{T}^2}\ , \quad (10)$$

which is independent of A and a function only of $\bar{\Omega}_E$ for a given μ^*. Denoting this function by $g(\bar{\Omega}_E,\mu^*)$ we have

$$\frac{\Delta C(T_c)}{\gamma(0)T_c}\equiv g(\bar{\Omega}_E,\mu^*)$$

so that a single curve applies for all values of A. The only approximation is the neglect of A in the Coulomb cutoff correction that can safely be ignored. Note that, in Fig. 2, we have used Ω_E/T_c instead of $\bar{\Omega}_E$, since it is easily shown that $\Delta C(T_c)/\gamma(0)T_c\equiv G(\Omega_E/T_c,\mu^*)$ with G an appropriate functional form.

V. RESULTS AND DISCUSSION

To check on the optimum spectrum (a delta function at Ω_E^* with entire weight A at a single frequency) we have worked out its functional derivative. This is shown in Fig. 2. The solid curve is

$$(1+\lambda)T_c\delta[\Delta C(T_c)/\langle\gamma(0)T_c\rangle]/\delta\alpha^2F(\omega)$$

(left-hand label) for a model delta function $\alpha^2F(\omega)=A\delta(\omega-\Omega_E^*)$ with Ω_E^* the frequency giving the maximum of the dashed curve. It is clear from the figure that the functional derivative is now very different from those found for realistic $\alpha^2F(\omega)$ spectra. It is negative definite with value zero right at Ω_E^*. This proves that a delta function at Ω_E^* gives a local maximum. Removal of some weight from the delta function at Ω_E^* and placing it at any other frequency reduces the specific-heat jump. Our experience with realistic spectra would lead us to believe that this is, in fact, an absolute maximum, although we have not found a rigorous mathematical proof. Figure 3 sheds more light on this point. In this figure we show as the solid line, the maximum value of $\Delta C(T_c)/\gamma(0)T_c$, obtained for different choices of μ^*. On the same figure, we show (dark points) values for the same quantity obtained in realistic cases.[12,14] They all fall below our theoretical local maximum indicating that for physical systems, the solid line is, indeed, an absolute maximum. Results of calculations based on Weber's spectrum[4] for the oxide $La_{1.85}Sr_{0.15}CuO_4$ give $\Delta C(T_c)/\gamma(0)T_c=2.82$, also below our maximum.

In an experiment on the specific-heat jump of the high-T_c oxide $La_{1.85}Sr_{0.15}CuO_4$ with $T_c=36$ K, Dunlap *et al.*[24] have found a value of $\Delta C(T_c)/\gamma(0)T_c$ of 2–10 depending on the analysis of the data. The upper limit obtained is higher than our theoretical maximum and, if confirmed, would rule out the electron-phonon interaction as the mechanism for this system. Having said this, it needs to be stressed that other measurements give more conservative values.[25–28] It is clear that accurate measurements of $\Delta C(T_c)/\gamma(0)T_c$ could make a critical contribution to our understanding of the mechanism responsible for the superconductivity in the oxides. Such accurate measurements are, however, very difficult. Not only is the jump a small correction over and above a large phonon specific-heat background, but the present value of $\gamma(0)$ is quite uncertain. As an example, when it is derived from a measurement of the slope of the upper critical field $H_{c2}(T)$ at T_c, quite different values of $\gamma(0)$ result if one uses a dirty or clean limit formula in the analysis. Also, the slope itself is uncertain. Orlando *et al.*[29] find -2.2 T/K using the midpoint of the transition and -5 T/K using the onset. This problem cannot be avoided until samples are produced, in which the resistive transition to the superconducting state is sharper.

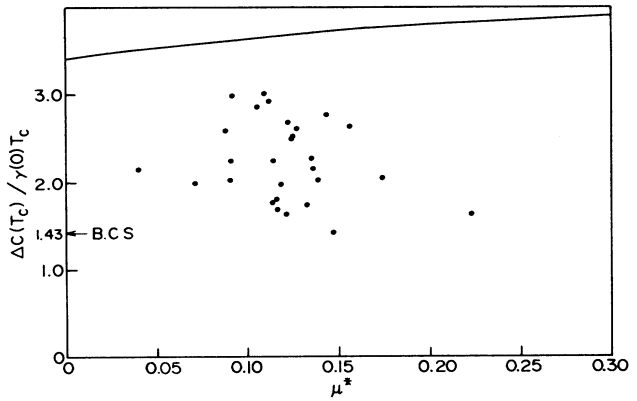

FIG. 3. The maximum possible value for $\Delta C(T_c)/[\gamma(0)T_c]$ as a function of μ^*. The solid dots represent theoretical values for the following materials in order of decreasing value of $\Delta C(T_c)/[\gamma(0)T_c]$: $Pb_{0.7}Bi_{0.3}$, $Pb_{0.65}Bi_{0.35}$, $Pb_{0.8}Bi_{0.2}$, $Pb_{0.9}Bi_{0.1}$, Pb, $Pb_{0.8}Tl_{0.2}$, Nb_3Sn, Nb_3Al, Nb_3Ge, $Pb_{0.6}Tl_{0.4}$, Hg, $Pb_{0.75}Bi_{0.25}$, $Pb_{0.4}Tl_{0.6}$, V_3Ga, $Pb_{0.5}Bi_{0.5}$, La, Ga (amorphous), Bi (amorphous), V_3Si, Mo (amorphous), Nb, In, $Tl_{0.9}Bi_{0.1}$, Tl, Sn, Ta, V, Al (BCS).

VI. CONCLUSIONS

In conclusion, we have found a local maximum for $\Delta C/\gamma(0)T_c$ which applies to electron-phonon superconductors for which the Eliashberg equations are assumed to be valid. We argue that this local maximum is probably an absolute maximum for realistic values of the electron-phonon spectral density $\alpha^2F(\omega)$. The maximum value of $\Delta C(T_c)/\gamma(0)T_c$ is surprisingly small and varies only slightly with the value of the Coulomb pseudopotential μ^*. As an example, it rises from a value of 3.4 for $\mu^*=0$ to a value of 3.9 for $\mu^*=0.3$. This last value is smaller than a recently suggested upper limit for the jump in $La_{1.85}Sr_{0.15}CuO_4$. Confirmation of this upper value as the actual value would rule out the electron-phonon mechanism in this material. Note that, since our maximum value for $\Delta C(T_c)/\gamma(0)T_c$ of 3.73 for $\mu^*=0.15$ is not that much larger than some of the experimental values found in real electron-phonon superconductors, it could be used to extract, from a measured value of $\Delta C(T_c)$, a lower limit on the zero-temperature Sommerfeld $\gamma(0)$. This may prove of some help in the analysis of experimental data.

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