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## Comments and Addenda

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# Superconducting lead: Quasiparticle recombination time and Kapitza resistance

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We deconvolve the system impulse response from the observed quasiparticle pulse transients of Hu *et al.* to determine the quasiparticle recombination time. Deconvolution of the accompanying phonon transient yields a phonon–helium-bath relaxation time from which we determine the Kapitza resistance of lead.

Hu *et al.* have described an experiment in which they create electron quasiparticles and phonons on one face of a superconducting lead crystal and detect, at the opposite face, the change with time of the local quasiparticle density, using as a detector a tunnel junction.[1] We find their analytical description of the data to be inadequate. Hence the conclusions which they draw from the data are without quantitative basis. We point out that the usual analytical description of such a system is in terms of two functions: (i) the space-time Green's function $G(r,r';t,t')$ appropriate for the particular sample boundary conditions, and (ii) the function which describes the generation of quasiparticles (QP) as a function of space and time $Q(r',t')$.[2-4] The density of QP at $r$ at time $t$, $n(r,t)$, is the space-time convolution of these two functions. We show that the sample Green's function at the detector is, with respect to the time variation of the observed QP density transient, a step function in time which then decays slowly at the rate at which QP are lost in the sample volume by recombination. This sample Green's function describes the observed transient signal at large times. The observed transient rise, with a step Green's function, is simply the time integral of the QP injection function $Q(r',t')$. We provide here an analysis which allows us to determine the quasiparticle recombination time in lead at 1.6 K to be 640 nsec.

Hu *et al.* use the Green's function for unbounded space alone to describe the observed transient in the QP density at the detector, $n(r,t)$. This Green's function is proportional to $t^{-3/2}\exp(-r^2/4Dt)$ for diffusive propagation of QP with a diffusion constant $D$, to a point $r$ from the source at a time $t$ following the injection of the impulse of QP. They point out that the tangent at the maximum slope of this function at a distance $L$ from the source intersects the time axis at $t_0=0.0375L^2/D$. Since they estimate $D$ from the residual resistance ratio to be $10^6$ cm$^2$/sec, by using this expression for $t_0$ their estimate of $D$ appears to be confirmed. However, we note that Hu *et al.* failed to point out that their infinite-medium Green's function reaches half-maximum for $L=0.87$ mm at $t_{1/2}=1.94t_0=0.6$ nsec, and the maximum is reached at $t_{\max}=4.44t_0=1.3$ nsec. The observed transient reaches these values at 40 and 120 nsec, some 70–100 times the times predicted by their improper Green's function. The function which they try to use to describe their signal therefore has no direct connection with their signal.

We proceed now to a presentation of a resumé of the canonical Green's-function description of this situation.[2-4] For completeness we discuss both diffusive and ballistic cases. First, we point out that for a finite sample volume the Green's function will be, essentially, a step function in time, regardless of the amount of diffusive scattering as long as some scattering takes place. The reason for this is simple. If in a short-time interval one creates $N$ quasiparticles at one point in the sample and these QP cannot be lost through the sample boundaries, then the mean QP density must increase by $N/V$, where $V$ is the sample volume, after some time comparable to the QP transit time in the sample. In the absence of any QP loss mechanism in the sample volume, such as QP recombination, the density would remain constant at this new level. With QP loss in the volume, the density will decay exponentially with a time constant $\tau_L$.

The Green's function for diffusive propagation in a sample in the form of a parallelopiped with sides $a$, $b$, $c$ has the form, if evaluated at a detector opposite an input point source,[2-4]

$$G(a,0:t,0) = \frac{N}{V} \sum^{\infty} \sum^{\infty} \sum^{\infty}_{k,m,n=-\infty} (-1)^k \times e^{-\pi^2 Dt\left(\frac{k^2}{a^2} + \frac{4m^2}{b^2} + \frac{4n^2}{c^2}\right)} . \tag{1}$$

This function for $a < b,c$ rises rapidly to a peak $(bc/3.4a^2)(N/V)$ at $t_{\max} = 1.6a^2/\pi^2 D \simeq 1.2$ nsec for $a = 0.87$ mm, and falls as $t^{-1}$ to the final density $N/V$. The relative importance in a convolution with a pulse of width $\tau$ of the spike to the step part of the Green's function is given approximately by $bct_{\max}/3.4a^2\tau$. In the present case the signal width $\tau$ appears to be 60 times $t_{\max}$ so the spike contribution is greatly reduced. Furthermore, the observed signal shows no obvious spike contribution. Hence, on the time scales of interest, the Green's function is a step function in time. A similar result is obtained with ballistic propagation.

The Green's function for a finite sample with ballistic propagation *and* scattering can be constructed in the same manner that the Green's function for the diffusive limit was generated, by an eigenfunction expansion, or by an image construction using the unbounded medium Green's function for ballistic propagation with scattering. We know that in the absence of scattering the Green's function for the unbounded medium is

$$\delta(|r - r'| - c(t - t'))/|r - r'| ,$$

where $c$ is the velocity of propagation. The Green's function for the finite sample, which is the sum of the images of this function, describes the arrival of the impulse excitation at a distance $L$ with a minimum delay which is $t_0 = L/c$. Without damping, the finite sample Green's function varies with time about the mean density which is the total number of particles in the impulse divided by the sample volume. In the experiment of Hu *et al.* the QP scattering by impurities is estimated to give a scattering time of $2 \times 10^{-10}$ sec, and the transit time is $8.7 \times 10^{-10}$ sec. It should be apparent, then, that the QP will propagate ballistically, and that the stationary QP density will be achieved in a few transit times. Alternatively, the impulse response for the ballistic system with scattering, which is known, can be used to determine the finite-sample Green's function. The fact that the diffusion constant $D$ is finite is certainly no criterion for evoking diffusive flow. The precise criterion, dictated by causality, is that the diffusive delay $t_0 = 0.048L^2/D$ must be equal to or greater than the ballistic delay $L/c$. For a given diffusion constant, a sample length $L_0$ will exist for which the propagation is ballistic. For greater lengths it will approach the diffusive limit. For the experiment of Hu *et al.*, this length is $L_0 = D/0.048c = 2$ mm. The conclusion, then, is that in all the crystals used by Hu *et al.*, which were 4.5 mm or shorter, the propagation transit time will be *indistinguishable* from $L/c$, that of a ballistic system. Hu *et al.* observed this to be the case, but they abandoned the observation and described the propagation as diffusive. The important effect of the finite diffusion constant which results from impurity scattering will be the blurring of the rise time of the sample impulse response rise time into a slower rise time typical of the pure diffusion response function, because of the delayed arrival of the scattered particles. Hence, with any assumed state of propagation, diffusive or ballistic, the Green's function will be a step function rising to the new QP density in a few nanoseconds.

Knowing that, on the time scales of interest, the Green's function is a step function $u(|r - r'| - c(t - t'))$ times an exponential with decay time $\tau_L$ to express the volume loss, the QP transient density expressed by the convolution

$$n(r,t) = \int_0^t \int_V Q(r',t') \times G(r,r':t,t')\, dr'^3\, dt' , \tag{2}$$

takes the simple form

$$n(r,t) = \int_V dr'^3 \int_{-\infty}^{t-|r-r'|/c} Q(r',t') \times e^{-(t-t')/\tau_L}\, dt' . \tag{3}$$

In general, however, Eq. (2) means that if one observes $N(r,t)$, and if one knows $G(r,r':t,t')$, then $Q(t)$ may be determined by deconvolution of Eq. (2) by any of several standard methods: by Laplace transforms, by matrix inversion, or by algebraic inversion if $G(r,r':t,t')$ obeys an addition theorem.[4]

In words, then, the rapidly rising and slowly falling parts of the QP transient signal can be understood in terms of the step-function character of the Green's function, and the convolution of Eq. (2). The QP density at the detector increases as the integral of the time variation of the QP input to the sample as the laser excitation of the sample turns on. After the laser turns off the QP injection diminishes. The integral of the flux, which represents the density of the QP at the detector, reaches a peak. It then slowly decays with a time constant which is the QP loss time constant $\tau_L$. The curve, Fig. 2 of Hu *et al.*, for 1.6 K shows a QP signal which may be represented by

$$n(L,t) = 1.5\bar{A}(1-e^{-t/32})^3 e^{-t/320} \ ,$$

where $\bar{A}$ is the maximum signal amplitude, and $t$ is in nsec. Hence one concludes that the single QP loss time constant $\tau_L$ is 320 nsec, or that the time constant for two-particle QP recombination $\tau_r$ is 640 nsec.[5] Kaplan *et al.* and Jaworski *et al.* show that one can use the experimentally determined coupling function for lead $\alpha^2F(\omega)$ to determine $\tau_r$ to be $2.0\times 10^{-12}T^{-1/2}e^{15.8/T}$.[5,6] This gives $\tau_r = 31$ nsec at 1.6 K. The disagreement is large.

If one carries out the convolution, one finds that, for a point source, the QP are injected in time as

$$Q(t) = 45\bar{A}(1-e^{-t/32})^2 e^{-t/29} \ .$$

This function describes the QP flux as having a rise time of 16 nsec, a decay of 32 nsec, and a half-amplitude full width of 64 nsec. These times are at least comparable with that of the laser input pulse having a rise time of 1.5 nsec, a width of 5 nsec, a sample-and-hold aperture of 10 nsec, and a diffusion width of 2.2 nsec. The QP transit time is 0.87 nsec. The contribution of the source size to the time width we have not sufficiently accurate data to estimate. QP must be created both by photoexcitation during the laser pulse and by thermal excitation during and after the laser pulse. The heating effect is important since $\tau_r$ decreases exponentially with increasing temperature. At 3 K, $\tau_r \sim 5$ nsec. Hence a small volume of the sample, heated to 3 K or above will rapidly produce a local QP density far above that of the sample equilibrium density.

Conventional thermal analysis shows that in cylindrical coordinates $(z,r,\theta)$, a heat flux $F(r,t)$ constant for a time interval $\tau$, and slowly varying radially, will, when injected into a surface at $z=0$, increase the sample temperature at the center of the heat flux as[3]

$$T \simeq [2F(0)/\kappa][f(t)-f(t-\tau)] \ ,$$

$$f(t) = (\alpha t/\pi)^{1/2}e^{-z^2/4\alpha t} + \tfrac{1}{2}z\,\mathrm{erf}[z/2(\alpha t)^{1/2}] \ , \qquad (4)$$

where $\kappa$ is the thermal conductivity, and $\alpha$ is the thermal diffusivity. At $z=0$, $t=\tau$, $T=2F(0)(\alpha\tau)^{1/2}/\kappa(\pi)^{1/2}$. At this time $\tau$ the sample will be heated appreciably to a depth $z=(4\alpha\tau)^{1/2}$. At this depth, $T=F(0)(\alpha\tau)^{1/2}(4\kappa)^{-1}$. For the lead sample of Hu *et al.* at 1.65 K, and without taking into account the variation of $\alpha$ and $\kappa$ from their values at that temperature, the heating depth is 0.04 mm, and the surface temperature is 2 K/(W absorbed). At $z=0.04$ mm, $T=0.44$ K/(W absorbed). Hu *et al.* used a 1-kW laser pulse, but only a small fraction of this was absorbed. At $4\tau=20$ nsec, these temperatures have decayed to 0.5 and 0.38 K/W. At $16\tau=80$ nsec, these temperatures are 0.22 and 0.2 K/W. The conclusion is that QP production will take place for times longer than the laser pulse length, just as the deconvolution of the transient data shows.

For the curves of Hu *et al.* taken at higher temperatures, we note that the two-particle relaxation times for 2.38, 2.6 and 2.95 K are $\tau_r = 21$, 11.2, and 5.1 nsec, if they are extrapolated from the $\tau_r$ which we have determined at 1.6 K. If one convolves the QP flux determined above with impulse responses having decays with half these times, one can show, after tedious but simple algebra, that the QP density, and hence the detector signal will be decreased by a factor of $e^{32/\tau_r}$. The predicted attenuation of the QP peak signal is thus 4.4, 18, and 500. The amplitude scale of the figure in Hu *et al.* is arbitrary, but these factors may be seen to describe the decline of the QP signal amplitude for the signals observed at 2.38, 2.6, and 2.95 K.

The data of Hu *et al.* taken at 2.95 K, where the QP recombination time is 5.1 nsec, shows no QP impulse peak, but it does show the change in the QP density at the arrival of the phonon heat pulse. The phonon heat-pulse rise-time is so long, the order of 600 nsec, that the QP density with a $\frac{1}{2}\tau_r = 2.5$-nsec relaxation time to the changing phonon density accurately follows the phonon density change in the vicinity of the QP detector as it changes with time. Here again we have the time variation of the phonon density observed at the detector being described as the convolution of the time dependence of the phonon excitation with the proper Green's function. The ballistic phonon impulse response will again be blurred by the diffusive scattering of phonons in the sample. The analysis of the phonon density transient, although complicated by the elastic anisotropy of lead and its three acoustical-mode branches is parallel to the analysis which we have given for the QP transient.

If one fits the data for 2.95 K in Fig. 1 of Hu *et al.* the detector signal is found to be

$$1.67\bar{A}(1-e^{-t/425})^3e^{-t/3570} \ ,$$

with time in nsec, and $\bar{A}$ is the peak of the detector output. Here the decay from the stationary phonon density, 3570 nsec, measures the rate at which thermal energy is lost to the helium environment.

By deconvolution the point-source phonon flux $P(t)$ is found to be

$$P(t) = 40\bar{A}(1-e^{-t/425})^2e^{-t/380} \ .$$

This function describes the heat as arriving with a rise time of 110 nsec, decaying in 350 nsec, and having a full width at half-amplitude of 560 nsec. The paraxial longitudinal- and transverse-phonon wave packets are not resolved because, as we have shown above, the source heat pulse is so much wider than 5 nsec. Their differing velocities would give a minimum width of $L(c_t^{-1} - c_l^{-1})$, where $L$ is the minimum transit length. For lead with a length of 0.87 mm in the [111] direction, this would be 425 nsec, a reasonable agreement with the observed width.

We have pointed out elsewhere that the observation of this decay of the phonon local density allows one to determine the Kapitza thermal resistance.[7] The relaxation time of the phonon density transient, 3.57 $\mu$sec, is equal to $CR_K V/S$, where $C$ is the specific-heat capacity of lead, $R_K$ is the Kapitza resistance, $V$ is the volume of the crystal, and $S$ is its surface area. Since $R_K$ is proportional to $T^{-3}$, the product $CR_K$ is nearly constant with change in temperature. Therefore all the phonon transients will be observed to decay with approximately the same time constant, as Hu *et al.* observed. In particular, at 2.95 K, we find $R_K = 0.6T^{-3}$ cm$^2$K/W. We write it in this form in order to compare this value, for a temperature above the helium lambda temperature, with experimental values measured below the lambda temperature. The experimental data are scattered, but the lowest resistance reported is $2.1T^{-3}$ cm$^2$K/W.[8] Our value is thus less than would be predicted from extrapolating other data, but it is still higher than the minimum given by the phonon-radiation limit, which for lead is $0.2T^{-3}$ cm$^2$K/W. Other heat leak paths, such as the sample suspension, will make the apparent $R_K$ less than the intrinsic $R_K$. In any case the exact number is not important here, since our main purpose is to point out the need and advantage of the deconvolution procedure in understanding heat-pulse data. To establish the magnitude of the Kapitza resistance with high confidence would require the carrying out of the experimental tests which we outline in Ref. 7.

As a final observation we note that in numerous heat pulse experiments reported in the past, the sample Green's function has not been deconvolved from the data, rendering their conclusions questionable.[9]

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