

### Comment on "Nonequilibrium Superconductivity: New Crystallographic and Magnetic Field Effects"

On the basis of charge-imbalance measurements on polycrystalline and single-crystal superconducting Nb, Johnson and Silsbee<sup>1</sup> (JS) suggest that the present ideas of quasiparticle diffusion and relaxation in superconductors should be reexamined. This Comment suggests that inhomogeneity in their Nb samples is a very serious problem; so serious that it casts considerable doubt on their conclusions concerning the validity of the present theory of charge imbalance in superconductors.

JS reason that their observations of the magnitude and temperature dependence of the detector voltage  $V_d$  (for fixed current injection) for both polycrystal and single-crystal Nb are in reasonable agreement with the present theory of charge imbalance, as formulated for homogeneous superconductors, and that the decay length found in polycrystalline Nb is also reasonable, so that it is surprising to find in single-crystal Nb an anomalous dependence of  $V_d(L, T)$  on distance  $L$  between the detector and injector corresponding to a very long diffusion length. From the close examination below, it appears that all of their data differ from expected behavior, not just the single-crystal data on  $V_d(L, T)$ , probably because of variations of  $T_c$  in each sample. The variations have a large effect very near  $T_c$ , where  $V_d(L)$  was measured, because the charge-imbalance decay length is expected theoretically to range from infinity in normal regions  $T/T_c \geq 1$  to about  $10 \mu\text{m}$  at  $T/T_c \approx 0.998$ , a range that corresponds to the observed 20 mK spread in  $T_c$ 's.

Let us compare the data with expected results. Qualitatively, the measured  $V_d(L, T)$  for fixed  $L$  shows several peaks as a function of temperature, whereas theory predicts a single peak diverging as  $(1 - T/T_c)^{-0.25}$  for  $T < T_c$ . JS ascribe the multiple peaks to a 10–20-mK variation in  $T_c$ . With this interpretation, reliable (non-zero) data lie below the peak in  $V_d$  at  $T - \bar{T}_c = -0.025$  mK, i.e., within the narrow 35-mK range  $-0.060$  mK  $< T - \bar{T}_c < -0.025$  mK. This restrictive criterion excludes all of the data on the decay length, i.e.,  $V_d(T, L)$ , which form the basis of the hypotheses of JS.

Quantitatively, from JS Fig. 2, the measured  $V_d(T, L)$  extrapolated to  $L = 0$ , i.e., detector and injector junctions coincident, is about 9 (4) times larger than expected for polycrystalline (single crystal) Nb. The discrepancy is inexplicably large in both cases. The estimates of 9 (4) use the model presented by JS, with their value  $\tau_0^0 = 25$  ps and with  $a = \frac{1}{2}$  as is appropriate for uniform diffusion of quasiparticles away from the injector junction. JS used  $a = \frac{3}{2}$ , which would be correct only if the diffusing quasiparticles were somehow restricted to the top third of the Nb bar.

Continuing the examination of the data on  $V_d(T, L)$ , temporarily overlooking the problem of inhomogeneity, I find another problem. From Eq. (1) of JS and  $\tau_0^0 = 25$  ps, the diffusion length  $\lambda_Q$  (20 mK) in polycrystalline Nb

is expected to be  $12 \mu\text{m}$  (with  $T_c = \bar{T}_c - 11$  mK), 3 times shorter than the fitted value of  $33 \mu\text{m}$  from  $V_d(L)$ . The discrepancy suggests two possible explanations:  $\tau_0^0$  in Nb is about 9 times larger than expected from the calculated electron-phonon scattering time for Nb, or the appropriate averaged  $T_c$  is about 1 mK above the temperature at which  $\lambda_Q$  was determined. The former explanation requires an implausible inaccuracy in the calculated time. (An enhanced rate would be more acceptable than an enhanced time, as is observed but not explained in the anomalously short charge-imbalance decay length in Al films.<sup>2,3</sup>) The latter explanation is inconsistent since it requires that the decay-length data be too close to  $T_c$  to be interpreted accurately, considering the 10–20-mK spread in  $T_c$ .

The measurements in a magnetic field are similarly difficult to interpret. Moreover, when a vortex lattice is induced by the field, the Nb may be dissipative at currents used in the injector junction, so that the spatial distribution of currents around the injector and detector junctions may change with temperature and/or field, thus affecting  $V_d$  in a complicated way not associated with charge imbalance.

Finally, it should be noted that the theory of charge-imbalance injection and detection in tunnel junctions on thin films recently has been confirmed to the 10% level in Sn and SnIn films,<sup>4</sup> both in magnitude and in temperature dependence, with good sample-to-sample reproducibility. The expected pair-breaking effects of applied supercurrents and supercurrents induced with a magnetic field have been verified in<sup>4</sup> Sn and SnIn and in<sup>5</sup> Al films. In consideration of this success, such a large failure of the theory as suggested by JS must be examined very carefully. Data are needed on more homogeneous materials over a wider temperature range before one concludes that the current theory is inadequate.

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