Comments and Addenda

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Specific heats of Li, Na, K, and Ag β-alumina below 1 K

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Specific-heat measurements of Li, Na, K, and Ag β-alumina in the temperature range 0.1-1 K confirm the existence of a broad spectrum of localized low-energy excitations.

Recently published[1] specific-heat data for M β-alumina (M = Li, Na, K, Rb, or Ag) over the range 1.3–40 K gave evidence of an excess specific heat at the lowest temperatures. It was surmised that the excess was due to a broad spectrum of localized low-energy excitations possibly arising from a tunneling motion of the metal cations within the conducting planes of the β-alumina. Evidence of these excitations has also been obtained from measurements of thermal conductivity[2] in the range 0.1–100 K and from mechanical damping[3] at kilohertz frequencies for temperatures above 1 K. To provide additional information about the densities of the low-energy excitations, we have extended the specific-heat measurements to 0.1 K for Li, Na, K, and Ag β-alumina.

The samples were prepared from 0.1–0.5 single crystals of melt-grown Na β-alumina[4] as discussed in Ref. 2. Measurements were made using a heat-pulse technique.[5] The calorimeter consisted of a 10^{-3}-cm-thick copper foil epoxied to the center of a stretched strip of 10^{-3}-cm-thick mylar film which provided a thermal link to the dilution refrigerator. Samples were attached to the copper foil with N-grease. A cut-down Matsushita carbon resistor[6] was used as a thermometer and Pt-W wire was used as a heater; both were epoxied to the Mylar opposite the copper foil. Electrical connections were made using multistranded, 7-μm diam superconducting Nb-Ti leads.[7] Temperatures were derived from a magnetic scale[8] calibrated against ^{3}He-vapor pressure and superconducting fixed points.[9] The heat capacity of the calorimeter was measured directly, allowing it to be subtracted from measurements when samples were attached. As a test, the specific heats of fused quartz and high-purity copper were measured. The results agreed with previous determinations[5,10] to 5%. As an additional test, a Ag β-alumina sample was remounted and rerun. The data from the two runs agreed to ≈ 2%. With the β-alumina samples in place, the addenda comprised roughly half the total heat capacity resulting in an estimated uncertainty of ≈ 15% in the magitude of the computed sample specific heats.

Our specific-heat data for M β-alumina are shown in Fig. 1, as are the lowest-temperature data ($T > 1.3$ K) taken from Ref. 1. The two sets of measurements agree reasonably well near 1 K, even though the samples of Ref. 1 were flux grown, whereas ours were melt grown.[11] The solid line in Fig. 1 represents the normal Debye contribution as calculated from acoustic phonon velocities.[12] Subtraction of the Debye contribution gives an excess specific heat of roughly 6.4 $T^{1.18}$, 7.3 $T^{1.22}$, 3.1 $T^{1.18}$, and 1.3 $T^{1.12}$ (in units of mJ/mol K) for

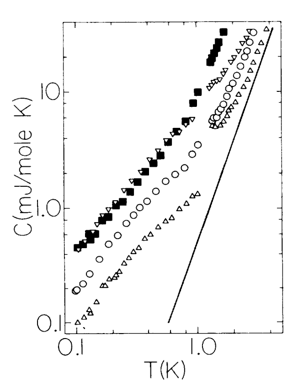

FIG. 1. Specific heat C of M β-alumina vs temperature. Data for $T \geq 1.3$ K were taken from Ref. 1. △, M = K; ○, Na; ■, Ag; ▽, Li. The solid line represents the Debye specific heat.

M = Ag, Li, Na, and K, respectively. The exponents are accurate to ≈5%. This clearly indicates the existence of a broad band of excitations extending to energies below 0.1 K, which is in agreement with the results from thermal conductivity measurements.[2]

If converted to specific heats per unit volume, the above values of excess specific heat are of the same temperature dependence and magnitude as observed for a variety of amorphous materials.[13] A similar statement applies to the thermal conductivity results.[2] Since both the specific heat and thermal conductivity are sensitive to the cation present, it is likely that the excitations are localized to the disordered[14] conducting planes. This would imply that these planes may be treated as a two-dimensional glass.

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[11]McWhan *et al.* of Ref. 1 did measure both a flux-grown and a melt-grown sample of Na β-alumina and observed little difference in specific heat. The samples of Ref. 1 were said to have about 26% excess cations, whereas ours contained 16–18%.

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[14]See, for example, P. D. Dernier and J. P. Remeika, J. Solid State Chem. 17, 245 (1976).