Charge Density Variation in $YBa_2Cu_3O_{6+y}$

In a recent Letter, Bobroff *et al.* [1] investigate the hole doping distribution and find that it is smaller than 0.025 based on ^{89}Y NMR. Their intent was to address inhomogeneous, nanoscale charge variations; however, their conclusions may appear to imply the absence of any type of charge-density modulation. While we agree with their data, we point out that their analysis does not rule out a largely commensurate charge-density variation of larger amplitude having the symmetry shown in Fig. 1.

The arrangement in Fig. 1 obeys the lattice symmetry, and is in agreement with the NMR observations (if the established hyperfine scenario is still valid): (1) The magnetic linewidths are all very small. (2) The electric field gradient distributions at Cu and O would be very narrow (^{89}Y is a spin $I = 1/2$ nucleus and thus not sensitive to charge variations). (3) One would expect there to be two oxygen sites that differ mainly in their electric field gradient, but *not* the magnetic shift. Indeed, as is known [2] from the early days of NMR, there are two nonequivalent oxygen sites that differ mostly in the electric field gradient. This splitting of the planar oxygen satellite lines has been attributed to the orthorhombic distortion [3], without there being proof. On the other hand, the experimentally observed splittings are not very different from the linewidths of the planar oxygen satellite transitions in $La_{2-x}Sr_xCuO_4$, where we proved the existence of charge-density variations [4] in the Cu-O plane (similar excessive widths have been reported in various other materials including $Tl_2Ba_2CuO_y$ [6]). Consequently, it cannot be excluded that the splittings observed in $YBa_2Cu_3O_{6+y}$ represent a commensurate charge-density variation in contrast to an incommensurate one in $La_{2-x}Sr_xCuO_4$, and the resulting (ordered) hole variation could exceed that estimated from ^{89}Y substantially. We also note that the possible charge and spin order is in agreement with the largely commensurate inelastic neutron scattering peaks found for $YBa_2Cu_3O_{6+y}$, as opposed to incommensurate peaks for $La_{2-x}Sr_xCuO_4$ [5].

To conclude, based on the NMR evidence given by Bobroff *et al.* [1] a large, but commensurate hole density variation in the Cu-O plane cannot be excluded.

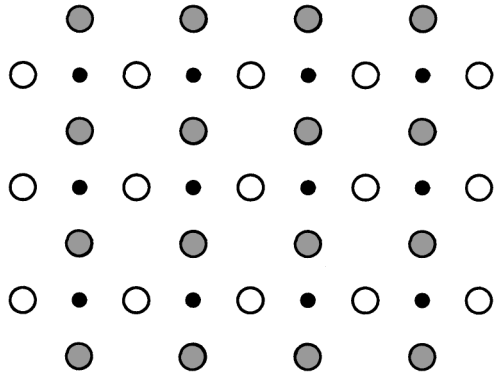

FIG. 1. Charge density variation in the Cu-O plane. The charge density at the planar oxygen sites (shaded and empty large circles) alternates when going around a Cu atom (small filled circle).

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Bobroff *et al.* Reply: In his Comment, Haase does not present a direct criticism of our own work but stresses that our study cannot rule out a commensurate charge distribution in the planes [1]. We never addressed such a possibility in our Letter, which was aimed at qualifying the *disorder*. We demonstrated that the hole content disorder, if any, is much smaller in YBCO than that estimated from the initial interpretation of the scanning tunneling microscopy data at the surface of Bi2212 samples [2]. The validity of our measurements, of their analysis, and of their significance is by no way disputed in the Comment by Haase.

He rather suggests the existence of a commensurate charge density variation in the CuO_2 planes which corresponds to a difference of charge between the two planar oxygens O(2) and O(3). This proposition addresses the interpretation of the ^{17}O NMR data of [3] in YBCO. In this study, a quadrupole splitting is observed between the two planar oxygen sites in $YBa_2Cu_3O_7$. This splitting is a proof that these two sites sense different electric field gradients (EFG). In [3], this splitting is interpreted to be due to the occurrence of the orthorhombic distortion associated with the existence of the filled CuO chains. The small observed splitting $\Delta\nu_Q/\nu_Q \approx 10\%$ is indeed compatible with a simple point charge model or more sophisticated models for the EFG [4]. However, Haase's proposition of a charge difference between O(2) and O(3) cannot be excluded, and is even expected as soon as an orthorhombic distortion occurs. In that case, it is hard to decide which effect drives the other.

The actual physical significance of Haase's proposition strongly depends on the magnitude of such a charge difference between O(2) and O(3). Haase advocates that this difference is *large,* but, in a recent detailed calculation, he finds only a relative variation of 9% of the charge on the oxygen sites [5]. We stress that this is in no way demonstrated in his Comment. One would need to separate the contribution to the EFG of the distant charges from that of the on-site charges to determine *quantitatively* the maximum charge unbalance between the two oxygen sites. We naively expect, as many others did before, that the charge unbalance is rather small as the EFG observed splitting does not exceed 10%. A thorough theoretical effort might allow a conclusion. Further experiments in other cuprates, especially nonorthorhombic ones such as the Tl or Hg compounds, would help as well to clarify this issue.

In our opinion, the significance of a *large* or *small* charge unbalance should not be purely semantic, but should refer to some specific physical effect. Experimentally this charge unbalance appears small for us as the system remains metallic in both *a* and *b* directions [6]. In such conditions we feel that Haase's proposition is not driving an essential property of the physics of the high T_C cuprates.

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