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LETTER TO THE EDITOR

Implications of the isotope effects on magnetization, magnetic torque and susceptibility

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Abstract

We analyse the magnetization, magnetic torque and susceptibility data of $La_{2-x}Sr_xCu^{16,18}O_4$ and $YBa_2^{63,65}Cu_3O_{7-\delta}$ near T_c in terms of the universal 3D-XY scaling relations. It is shown that the isotope effect on T_c mirrors that on the anisotropy γ. Invoking the generic behaviour of γ, the doping dependence of the isotope effects on the critical properties, including T_c, correlation lengths and magnetic penetration depths, are traced back to a change of the mobile carrier concentration.

(Some figures in this article are in colour only in the electronic version)

A lot of measurements of the oxygen and copper isotope effect on magnetization, susceptibility, magnetic torque etc have been performed on a variety of cuprate superconductors [1–9]. To obtain estimates for the shift of the transition temperature T_c upon isotope exchange, the respective mean-field models [10, 11] have been invoked. For the magnetization m they imply that the mean-field transition temperature T_{c0} can be estimated by extrapolating $m(T)$ linearly. To illustrate this traditional treatment we display in figure 1 the data of Batlogg *et al* [1] for a nearly optimally doped $La_{1.85}Sr_{0.15}Cu^{16,18}O_4$ powder sample in terms of $^{16,18}m(T)$ versus T. The straight lines are parallel linear extrapolations, yielding the mean-field estimates $^{16}T_{c0} \approx 34.56$ K, $^{18}T_{c0} \approx 34.12$ K and $\Delta T_{c0}/T_{c0} \approx -0.012$, where $\Delta Y = (^{18}Y - ^{16}Y)/^{18}Y$. However, the reliability of these estimates is doubtful because the dominant role of critical fluctuations near T_c [12–21] and the isotope effects on the penetration depth [22–25] etc are neglected, the anisotropy is not taken into account, and the choice of a linear and parallel portion appears arbitrary.

Noting that the isotope effects in cuprate superconductors pose a fundamental challenge in their understanding, the shortcomings of the traditional interpretation of magnetization and related data call for a treatment that goes beyond mean-field and takes the anisotropy into account. In this work we concentrate on the critical regime of anisotropic extreme type II superconductors where 3D-XY fluctuations dominate [12–21]. Invoking the universal

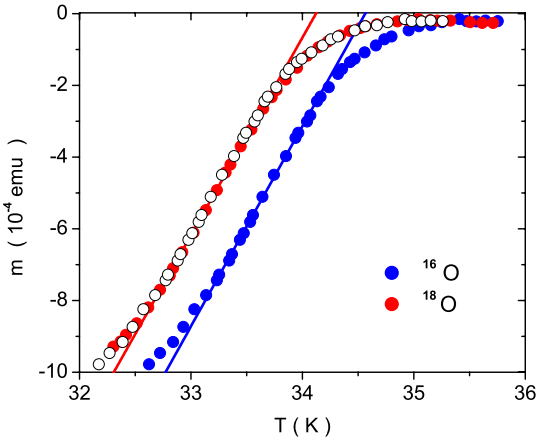

Figure 1. $^{16,18}m(T)$ versus T for a $La_{1.85}Sr_{0.15}Cu^{16,18}O_4$ powder sample at 6.5 Oe taken from Batlogg *et al* [1]. The parallel straight lines are linear extrapolations, yielding the mean-field estimates $^{16}T_{c0} \approx 34.56$ K, $^{18}T_{c0} \approx 34.12$ K and $\Delta T_{c0}/T_{c0} \approx -0.012$. The black circles are the $^{16}m(T)$ data rescaled according to equation (4) with $a \simeq 0.986$.

scaling functions for magnetization and magnetic torque we analyse the magnetization data of $La_{1.85}Sr_{0.15}Cu^{16,18}O_4$ [1], the magnetic torque data of $La_{1.92}Sr_{0.08}Cu^{16,18}O_4$ [6] and the susceptibility data of $YBa_2^{63,65}Cu_3O_{7-\delta}$ [4] near T_c. An essential additional relation emerges from the observation that data taken at fixed magnetic field and on samples with ^{16}O or ^{18}O, as well as with ^{63}Cu or ^{65}Cu, collapse near criticality within experimental error, when the temperature is rescaled appropriately. Although this property provides retrospectively partial support for the traditional approach [1–9], 3D-XY scaling uncovers the essential role of the anisotropy γ. Indeed, the change of T_c is found to mirror the shift of the anisotropy γ. As a consequence, the generic shift of the temperature dependent magnetization, susceptibility and magnetic torque upon isotope exchange at fixed magnetic field does not provide estimates for the change of the transition temperature only, as hitherto assumed [1–9]. Together with the generic behaviour of the anisotropy [18, 26–31], the doping dependences of the isotope effects are then traced back to the change Δx_u of the underdoped limit x_u. This implies a shift of the phase diagram in the temperature-doping plane towards a slightly higher dopant concentration x, along with a reduction of the mobile charge carrier concentration $\overline{\delta} = x - x_u$. This contribution leads to a negative shift of T_c. We identify a positive shift as well. This stems from the change $\Delta\gamma_0$ of the critical amplitude γ_0 at the quantum superconductor to insulator transition. The magnitude and proportion of these contributions is controlled by Δx_u and $\Delta\gamma_0$. Their values are material dependent. In any case, they control the isotope effects and remain to be understood microscopically. However, the emerging essential role of the anisotropy represents a serious problem for two-dimensional models as candidates to explain superconductivity in the cuprates, and serves as a constraint on future work towards a complete understanding.

Whenever 3D-XY fluctuations dominate the magnetization, m adopts the scaling form [13–21]

$$\frac{m(T,\delta,H)}{T\sqrt{H}} = -\frac{k_B Q_3}{\Phi_0^{3/2}}\gamma\epsilon^{3/2}(\delta)\frac{1}{\sqrt{z}}\frac{dG(z)}{dz}, \qquad z = \frac{H\xi_{ab}^2}{\Phi_0}\epsilon(\delta), \tag{1}$$

where $\epsilon(\delta) = (\cos^2(\delta) + \sin^2(\delta)/\gamma^2)^{1/2}$ and $\gamma = \xi_{ab}/\xi_c$ denotes the anisotropy. Q_3 is a universal constant, $G(z)$ a universal function of its argument, $\xi_{ab,c}$ the correlation lengths

in the ab-plane and along the c-axis, H the magnetic field and Φ_0 the flux quantum. Close to the zero-field transition temperature T_c the correlation lengths diverge as $\xi_{ab,c} = \xi_{ab0,c0}|t|^{-\nu}$, where $\nu \simeq 2/3$ and $t = T/T_c - 1$. An essential implication is that in the plot $m(T, \delta, H)/(\gamma\epsilon^{3/2}(\delta)T\sqrt{H})$ versus $z = (H\xi_{ab0}^2\epsilon(\delta)/\Phi_0)|t|^{-2\nu}$ or $x = z^{-1/2\nu} = (\Phi_0/(H\xi_{ab0}^2\epsilon(\delta)))^{1/2\nu}t$ the data fall close to criticality on a single curve. For $YBa_2Cu_3O_{7-\delta}$ this scaling property is experimentally well confirmed [14, 16]. Because the magnetization exists at T_c the combination $m(T, \delta, H)/(\gamma\epsilon^{3/2}(\delta)T\sqrt{H})$ adopts the universal value [15, 17–20]

$$\frac{m(T_c)}{\gamma(T_c)\epsilon^{3/2}(\delta)T_c\sqrt{H}} = -\frac{k_B Q_3 c_{3,\infty}}{\Phi_0^{3/2}}, \tag{2}$$

where $c_{3,\infty}$ is a universal constant [15, 17–20]. Thus, plotting $m(T)/(\gamma\epsilon^{3/2}(\delta)\sqrt{H})$ versus T, the data taken in different fields cross at T_c. In powder samples and sufficiently large anisotropy ($\gamma \gg 1$) this relation reduces to

$$\frac{m(T_c)}{\gamma(T_c)T_c\sqrt{H}} = -\frac{\pi k_B Q_3 c_{3,\infty}\langle|\cos(\delta)|^{3/2}\rangle}{2\Phi_0^{3/2}}. \tag{3}$$

As the oxygen isotope effect on the magnetization at fixed magnetic field is concerned it implies that the relative shifts of magnetization m, anisotropy γ and T_c are not independent but related by $\Delta m(T_c)/m(T_c)+\Delta\gamma(T_c)/\gamma(T_c)+\Delta T_c/\Delta T_c = 0$. On that condition it appears impossible to extract T_c and ΔT_c from the temperature dependence of the magnetization taken in one particular magnetic field. However, there is the special case where close to criticality the data scale as

$$^{18}m(T) = {}^{16}m(aT), \tag{4}$$

within experimental error. Subsequently it would also justify the traditional method of extracting $\Delta T_c/T_c$ [1–9]. A glance at figure 1 shows that this scale transformation is well confirmed within experimental error, roughly given by the size of the symbols of the data points. It yields $\Delta m(T_c)/m(T_c) \simeq 0$ with $a = {}^{18}T_c/{}^{16}T_c \simeq 0.986$. Thus, $\Delta T_c/T_c \simeq -\Delta\gamma(T_c)/\gamma(T_c) \simeq -0.014$ for $La_{1.85}Sr_{0.15}Cu^{16,18}O_4$. Although $\Delta T_c/T_c \simeq -0.014$ is close to the traditional estimate, $\Delta T_{c0}/T_{c0} \approx -0.012$, its significance is fundamentally different. Indeed, because $\Delta m(T_c)/m(T_c) \simeq 0$, the universal relation $\Delta m(T_c)/m(T_c) + \Delta\gamma(T_c)/\gamma(T_c) + \Delta T_c/\Delta T_c = 0$ reduces to

$$-\frac{\Delta T_c}{T_c} = \frac{\Delta\gamma(T_c)}{\gamma(T_c)} = \frac{\Delta\xi_{ab0}}{\xi_{ab0}} - \frac{\Delta\xi_{c0}}{\xi_{c0}} = 1 - a. \tag{5}$$

Hence, the isotope effect on T_c mirrors that on the anisotropy and the critical amplitudes of the correlation lengths. In virtue of the universal scaling expression (2), relation (4) is obtained when the anisotropy γ scales near T_c as $\gamma(T) = a\gamma(aT)$. The rescaled curves should then cross at T_c, provided that the experimental uncertainties do not mask the isotope induced change of ξ_{ab0}, the critical amplitude of the in-plane correlation length. Considering the data shown in figure 1 we observe that the two curves coincide in the critical regime within experimental error, roughly given by the size of the symbols of the data points. Apparently, the resolution of the crossing point requires considerably more accurate data. Otherwise, as in the present case, the coincidence of the rescaled data confirms the consistency with 3D-XY critical behaviour and allows us to determine the rescaling factor a around T_c, and with equation (5) to estimate the shifts $\Delta T_c/T_c$ and $\Delta\gamma(T_c)/\gamma(T_c)$ rather accurately.

Before turning to the implications of these results, revealing that the isotope effect on T_c mirrors that on the anisotropy γ, it is essential to explore the effect of the dopant concentration. Since sufficiently dense data only appear to be available for underdoped $La_{1.92}Sr_{0.08}Cu^{16,18}O_4$, we are left with the reversible magnetic torque data of Hofer *et al* [6] shown in figure 2 in terms of τ versus T. At T_c, fixed orientation and magnitude of the applied field τ scales

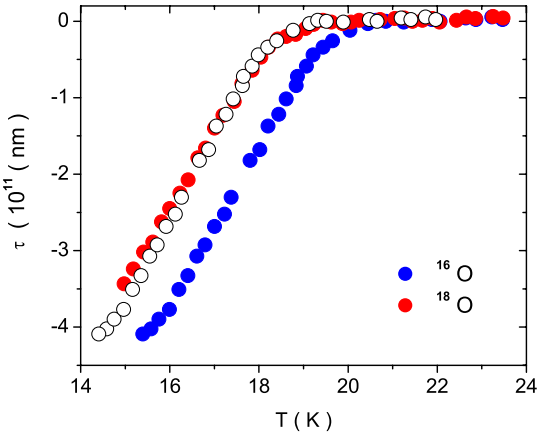

Figure 2. Reversible magnetic torque $\tau(\delta)$ versus T at $H = 0.1$ T and $\delta = 45°$ for $La_{1.92}Sr_{0.08}Cu^{16,18}O_4$ taken from Hofer *et al* [6]. The black circles are ${}^{18}m(T) \simeq {}^{16}m(aT)$ with $a = 0.936$.

as $\tau(T_c) = -\text{const} \times T_c\gamma(T_c)H^{3/2}$ [15, 17–20] and at fixed magnetic field the shifts are related by $\Delta\tau(T_c)/\tau(T_c) + \Delta\gamma(T_c)/\gamma(T_c) + \Delta T_c/\Delta T_c = 0$. From figure 2 it is seen that with ${}^{18}\tau(T) = {}^{16}\tau(aT)$ and $a = 0.936$ near coincidence is achieved within experimental accuracy. Since the universal scaling law for the magnetic torque [15, 17–19] is essentially analogous to equation (2) the near coincidence implies $\Delta\tau(T_c)/\tau(T_c) \simeq 0$ and (5) holds in this case as well, so that $\Delta T_c/T_c \simeq -\Delta\gamma(T_c)/\gamma(T_c) \simeq -0.07$.

To check the generic significance of this scenario we also analysed the susceptibility data for the copper isotope effect in $YBa_2^{63,65}Cu_3O_{7-\delta}$ of Zhao *et al* [4]. For all four dopant concentrations, extending from the underdoped to the optimally doped regime, we find that ${}^{65}\chi(T) = {}^{63}\chi(aT)$ is satisfied within experimental error, as discussed below. Since $\chi = m/H$ this strongly suggests that the scaling relation (5) holds for both copper and oxygen isotope exchange, irrespective of the doping level.

Having established the consistency with 3D-XY critical behaviour, together with the experimental facts that at T_c and fixed magnetic field $\Delta m/m$, $\Delta\tau/\tau$ and $\Delta\chi/\chi$ vanish within experimental error, the isotope effect on T_c does not only mirror that of the anisotropy (equation (5)), but is also subject to the other universal relations of the 3D-XY universality class. In particular, T_c, ξ_{c0} and λ_{ab0} are not independent but related by the universal relation [13, 15, 19, 20, 23, 32] $T_c = B\xi_{c0}/\lambda_{ab0}^2$, where B is a universal constant and λ_{ab0} the critical amplitude of the in-plane penetration depth. This leads for the respective relative shifts upon isotope exchange to the additional relation $\Delta T_c/T_c = \Delta\xi_{c0}/\xi_{c0} - 2\Delta\lambda_{ab0}/\lambda_{ab0}$. The lesson is, whenever 3D-XY fluctuations dominate, the isotope effects, e.g. on T_c,γ and λ_{ab0}, are not independent. These relations are particularly useful to open a door towards the understanding of the common origin of these effects. For example, given a generic relationship between anisotropy γ and mobile carrier concentration $\tilde{\delta} = x - x_u$ at fixed x, where x_u is the underdoped limit, the isotope effects in the cuprates would arise from a shift of x_u and the associated change of $\tilde{\delta}$. As shown in figure 3 for $La_{2-x}Sr_xCuO_4$, the generic doping dependence of γ is well established in a rich variety of cuprates in terms of the empirical relation [20, 31, 33]

$$\gamma(T_c) = \frac{\gamma_0}{\tilde{\delta}} = \frac{\gamma_0}{x - x_u}, \tag{6}$$

where γ_0 is a material dependent constant. Approaching the underdoped limit, where the cuprates correspond to an independent stack of sheets with thickness d_s [20, 31, 33], this

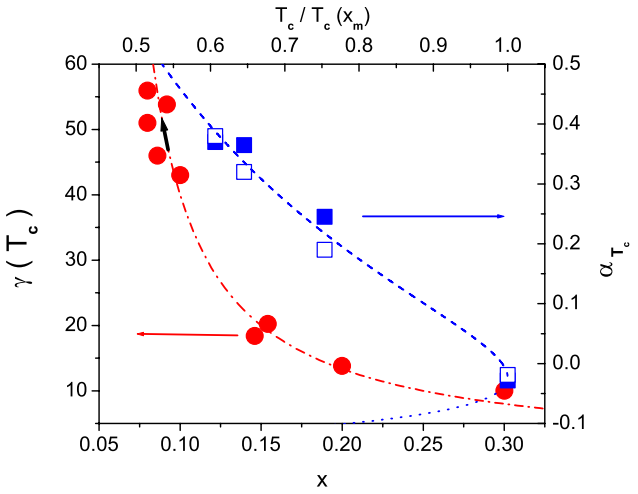

Figure 3. $\gamma(T_c)$ versus x for $La_{2-x}Sr_xCuO_4$ taken from [18, 26–30] (●) and α_{T_c} versus $T_c/T_c(x_m)$ for $YBa_2^{63,65}Cu_3O_{7-\delta}$ taken from Zhao *et al* [4] (■). The dash–dot line is equation (6) with $x_u = 0.05$ and $\gamma_0 = 2$. The arrow indicates the flow to the superconductor to insulator transition. The open squares result from the scaling analysis of the susceptibility data for the samples with $7-\delta = 6.94, 6.75, 6.63$ and 6.48 in terms of ${}^{65}\chi(T) = {}^{63}\chi(aT)$, yielding $a \simeq 1.0006, 0.994, 0.990$ and 0.988, respectively. The dashed and dotted curves result from equations (7) and (8) with $\Delta\gamma_0/\gamma_0 = -0.0082$, $\Delta x_u = 0.0012$ and $T_c(x_m) = 92.37$ K.

relation follows from the doping dependence of the critical amplitudes of the correlation lengths. Since ξ_{c0} tends to d_s, while ξ_{ab0} diverges as $\xi_{ab0} = \overline{\xi_{ab0}}/(x - x_u)$ [20, 31, 33] we obtain $\gamma_0 = d_s/\overline{\xi_{ab0}}$, which is the critical amplitude of the anisotropy at the quantum superconductor to insulator transition at x_u. The doping dependence of the relative isotope shifts is then traced back to a change of γ_0 and the shift Δx_u of the underdoped limit and with that to a change of the mobile carrier concentration $\tilde{\delta} = x - x_u$ according to

$$\frac{\Delta\gamma(T_c)}{\gamma(T_c)} = \frac{\Delta\gamma_0}{\gamma_0} + \frac{\Delta x_u}{\tilde{\delta}} = \frac{\Delta\gamma_0}{\gamma_0} + \frac{\Delta x_u}{x_m\left(1 \pm \sqrt{1 - T_c/T_c(x_m)}\right)}, \tag{7}$$

where we invoked the empirical relation between the hole concentration x and T_c due to Presland *et al* [34]. $x_m \simeq 0.16$ denotes optimum doping. This leads to the important conclusion that the doping dependence of the isotope effects in the cuprates stems from the shift of the underdoped limit. Finally, combined with equation (5) we obtain

$$\frac{\Delta\gamma(T_c)}{\gamma(T_c)} = \frac{\Delta\gamma_0}{\gamma_0} + \frac{\Delta x_u}{\tilde{\delta}} = 1 - a = -\frac{\Delta T_c}{T_c} = \frac{\Delta\xi_{ab0}}{\xi_{ab0}} - \frac{\Delta\xi_{c0}}{\xi_{c0}} = -\frac{\Delta\xi_{c0}}{\xi_{c0}} + \frac{2\Delta\lambda_{ab0}}{\lambda_{ab0}}, \tag{8}$$

relating the various relative shifts to the scaling factor a. With our estimates $\Delta T_c/T_c \simeq -0.014$ ($x = 0.15$) and $\Delta T_c/T_c \simeq -0.07$ ($x = 0.08$) for $La_{2-x}Sr_xCuO_4$ and relations (7) and (8) we obtain $x_u = 0.05$ for the essential, but material dependent, parameters, determining the doping and T_c dependence of $\alpha_{T_c} = -(M/\Delta M)\Delta T_c/T_c$ and the values $\Delta\gamma_0/\gamma_0 \simeq -0.01$ and $\Delta x_u \simeq 0.0024$. To illustrate this feature and to check the generic significance of this scenario further we consider the copper isotope effect on T_c in $YBa_2Cu_3O_{7-\delta}$. As aforementioned, our scaling analysis of the susceptibility data of Zhao *et al* [4] reveals full consistency with ${}^{65}\chi(T) = {}^{63}\chi(aT)$ for all doping concentrations within experimental error. Since $\chi = m/H$ the implications are equivalent to those derived for the magnetization and equation (8) applies as well. The resulting estimates for $\alpha_{T_c} = -(M/\Delta M)\Delta T_c/T_c$ are included in figure 3 and

compared with those obtained from the traditional extrapolation approach [4]. More importantly, given $\Delta\gamma_0/\gamma_0$ and Δx_u the T_c dependence of α_{T_c} is readily calculated with the aid of equations (7) and (8). As shown in figure 3 in terms of the dashed line, agreement is achieved with $\Delta\gamma_0/\gamma_0 \simeq -0.0082$ and $\Delta x_u \simeq 0.0012$. In comparison with $Y_{1-y}Pr_yBa_2Cu_3{}^{16,18}O_{7-\delta}$ the data of Franck *et al* [35] yields $\Delta\gamma_0/\gamma_0 \simeq -0.0060$ and $\Delta x_u \simeq 0.0019$.

In summary, we have seen that near T_c, where 3D-XY fluctuations are essential, the isotope effects on various critical properties are not independent but related by universal relations. Together with the observation that data taken at fixed magnetic field and on samples with ^{16}O or ^{18}O, as well as with ^{63}Cu or ^{65}Cu, collapse near criticality within experimental error, when the temperature is rescaled appropriately, we derived an additional relationship. It reveals the essential relevance of the anisotropy γ. Indeed, the relative shift of T_c was found to mirror that of the anisotropy γ. As a consequence, the temperature shift of the magnetization, susceptibility and the magnetic torque at fixed magnetic field does not only provide estimates for the change of the transition temperature, as hitherto assumed [1–9]. Together with the generic behaviour of the anisotropy [18, 26–30], the doping dependence of the isotope effects was traced back to a change of the underdoped limit Δx_u, or in other words to a shift of the phase diagram in the temperature–doping plane towards a slightly higher dopant concentration, along with a reduction of the mobile charge carrier concentration. This contribution leads to a negative shift of T_c. We identified a positive shift as well. It stems from the change of the critical amplitude of the anisotropy γ_0 at the quantum superconductor to insulator transition. The magnitude and proportion of these contributions is controlled by Δx_u and $\Delta\gamma_0$. Their values turned out to be material dependent. In any case, they control the isotope effects and remain to be understood microscopically. However, the emerging essential role of the anisotropy represents a serious problem for two-dimensional models as candidates to explain superconductivity in the cuprates, and serves as a constraint on future work towards a complete understanding. In addition, isotope exchange leads unavoidably to lattice distortions. The coupling with the in-plane penetration depth was recently established [24].

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