

# Onset of dielectric modes at 110 K and 60 K due to local lattice distortions in nonsuperconducting $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ crystals

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We report the observation of two dielectric transitions at 110 and 60 K in the microwave response of nonsuperconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  crystals. The transitions are characterized by a change in polarizability and presence of loss peaks, associated with overdamped dielectric modes. An explanation is presented in terms of changes in polarizability of the apical O atoms in the Ba–O layer, affected by lattice softening at 110 K, due to change in buckling of the Cu–O layer. The onset of another mode at 60 K strongly suggests an additional local lattice change at this temperature. Thus microwave dielectric measurements are sensitive indicators of lattice softening which may be relevant to superconductivity.

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It was recognized soon after the discovery of the high-temperature superconductors that the cuprates are structurally similar to the ferroelectric perovskites.<sup>1</sup> The basic perovskite  $\text{ABO}_3$  structure occurs in ferroelectrics like  $\text{BaTiO}_3$  and incipient ferroelectrics or quantum paraelectrics such as  $\text{SrTiO}_3$ , as well as in subunits of the superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0+x}$ . The implications of this structural similarity received early support from the observation of large dielectric response<sup>2,3</sup> in the insulating parent compound  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ . Furthermore, theoretical models have been proposed which include the possible competition between ferroelectricity and superconductivity.<sup>4–6</sup>

In this paper we show some striking dielectric properties of single crystals of insulating  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  which seem to have a strong bearing on the superconductivity of doped  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0+x}$ . Traditionally the information on lattice dynamics has been obtained from inelastic neutron, x-ray absorption fine structure (XAFS), Raman, and infrared spectroscopy measurements. Our microwave measurements probe consequences of lattice modes on the long wavelength ( $q=0$ ) dielectric properties, and its very high sensitivity leads to the observation of features not detected by other techniques.

In addition to large dielectric strengths  $\epsilon' \sim 10^2 - 10^3$ , consistent with previous measurements, we report the presence of two dielectric transitions at 110 and 60 K. These transitions are accompanied by the onset of polarization modes indicated by the presence of dielectric loss peaks below the transition temperatures. The transitions arise from structural distortions occurring at these temperatures, such as buckling of the Cu–O plane leading to the 110 K transition, which affect the electrodynamic response. Thus precision microwave measurements are shown to be a sensitive probe of lattice effects, complementing other traditional probes of lattice dynamics. Taken together with numerous reports of lattice effects at or near the superconducting transition temperature  $T_c$ ,<sup>7</sup> the present results demonstrate the importance of charge and lattice dynamics in the high-temperature superconducting oxides.

Ultrapure single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  were prepared in contamination-free  $\text{BaZrO}_3$  crucibles. The high quality of these single crystals has been extensively documented in a wide range of measurements, including structural and transport studies. A brief list of these reports can be found in Ref. 8.

The high sensitivity microwave measurements were carried out in a Nb superconducting cavity resonant at 10 GHz in the  $TE_{011}$  mode. The sample is placed at the center of the cavity at a maximum of the microwave magnetic field  $H_\omega$ . We introduce an electromagnetic susceptibility  $\tilde{\zeta}_H(T) = \zeta'_H(T) + i\zeta''_H(T)$  which is related to the measured parameters, the shift in cavity resonant frequency  $\delta f(T)$  and the resonance width  $\Delta f(T)$  by  $\delta f(T) - i\Delta f(T) = -g[\zeta'_H(T) + i\zeta''_H(T)]$ , where  $g$  is a geometric factor. A detailed analysis of the relevant cavity perturbation for general sample conditions including lossy dielectric and metallic or superconducting states, has been recently carried out by us.<sup>9</sup> We are able to directly measure the conductivity  $\tilde{\sigma}_{tot}$  or the dielectric permittivity  $\tilde{\epsilon}_{tot}$  (where  $\tilde{\sigma}_{tot} = -i\omega\epsilon_0\tilde{\epsilon}_{tot}$ ). The analysis shows that for arbitrary conductivity,

$$\tilde{\zeta}_H(T) = -\frac{3}{2} [1 - 3\tilde{z}^2 + 3 \cot(\tilde{z})/\tilde{z}], \quad (1)$$

$$\text{where } \tilde{z} = ka = k_0 a \sqrt{\tilde{\epsilon}_{tot}}.$$

Note that we use time dependences  $e^{-i\omega t}$ . In the limit  $\tilde{z} \ll 1$ ,  $\tilde{\zeta}_H(T) \approx (1/10)(k_0 a)^2(\tilde{\epsilon}_{tot} - 1)$  for a lossy dielectric. The dielectric permittivity  $\tilde{\epsilon}_{tot}$  was extracted from the data using Eq. (1).  $\tilde{\epsilon}_{tot}$  includes both bound polarization ( $\tilde{\epsilon}$ ) and free charge conductivity  $\tilde{\sigma}$  contributions, i.e.,  $\tilde{\epsilon}_{tot} = \tilde{\epsilon} + i\tilde{\sigma}/\omega\epsilon_0$ . In the present case the conductivity is negligible and  $\tilde{\epsilon}_{tot} = \tilde{\epsilon}$ . We have earlier carried out extensive measurements of the surface impedance of a variety of superconductors,<sup>10</sup> metals, and insulators, and demonstrated the validity of these measurements.

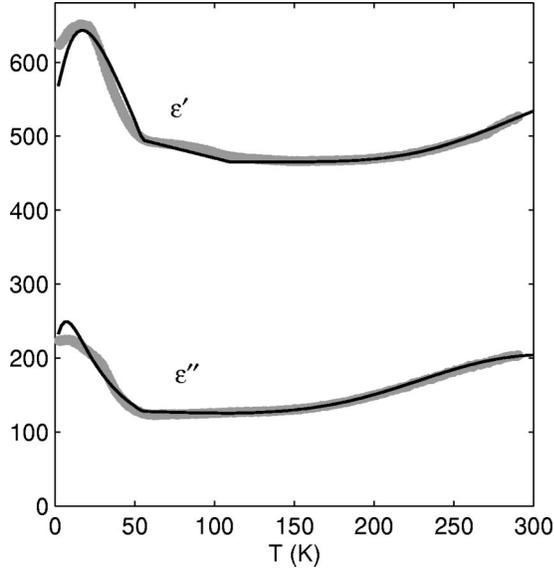


FIG. 1. Dielectric constants  $\epsilon'$  and  $\epsilon''$  at 10 GHz of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0+x}$ . Note the onset of dielectric response at  $T_{cA}=60$  K and  $T_{cB}=110$  K. The solid lines represent  $\epsilon = \epsilon_\alpha + \epsilon_\beta + \epsilon_\gamma$  as described in the text.

The dielectric permittivity  $\epsilon'(T)$  and  $\epsilon''(T)$  of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  are shown in Fig. 1. Here  $H_\omega \parallel \hat{c}$  axis, so that the displacement currents are in the  $ab$  plane, i.e., we are measuring in-plane dielectric permittivity  $\tilde{\epsilon}_{ab}$ . The large microwave dielectric permittivity observed in the present composition seems to be a characteristic of some perovskite oxides. Such large dielectric strengths  $\epsilon' \sim 10^2 - 10^3$  in nonmetallic insulating  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0+x}$  were reported by Rey *et al.*,<sup>2</sup> for ceramic samples which were quenched to retain the oxygen homogeneity. It is worth remarking that the present crystals are also quenched from high temperature and this may be an important requirement for the observation of this effect. We have observed similar response in the microwave dielectric permittivity of another  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  crystal (Fig. 2, bottom panel) obtained from a different batch, confirming the presence of the dielectric transitions reported here.

The data in Fig. 1 can be analyzed in terms of three dielectric modes,  $\tilde{\epsilon} = \tilde{\epsilon}_\alpha + \tilde{\epsilon}_\beta + \tilde{\epsilon}_\gamma$ , each of which is well described by a Debye relaxation form with respect to the temperature dependence:

$$\tilde{\epsilon} = \tilde{\epsilon}_\alpha + \tilde{\epsilon}_\beta + \tilde{\epsilon}_\gamma = \sum_{i=\alpha,\beta,\gamma} \frac{\epsilon_{i0}(T)}{1 - i\omega\tau_i(T)}. \quad (2)$$

$\tilde{\epsilon}_\gamma$  appears to represent the low  $T$  tail of a high-temperature process, with  $\epsilon_{\gamma 0}(T) = 160$ , and with a relaxation time  $\tau_\gamma(T) = 6.5 \times 10^{-13} \text{ sec}^{-1} \exp(1000/T)$  characterized by an activation energy of 1000 K. The  $\tilde{\epsilon}_\gamma$  process is dominant between 300 K and approximately 180 K, below which it “freezes” out quasistatically as the dipole relaxation rate becomes extremely slow. A residual temperature independent dielectric contribution  $\approx 465 + i125$  remains at

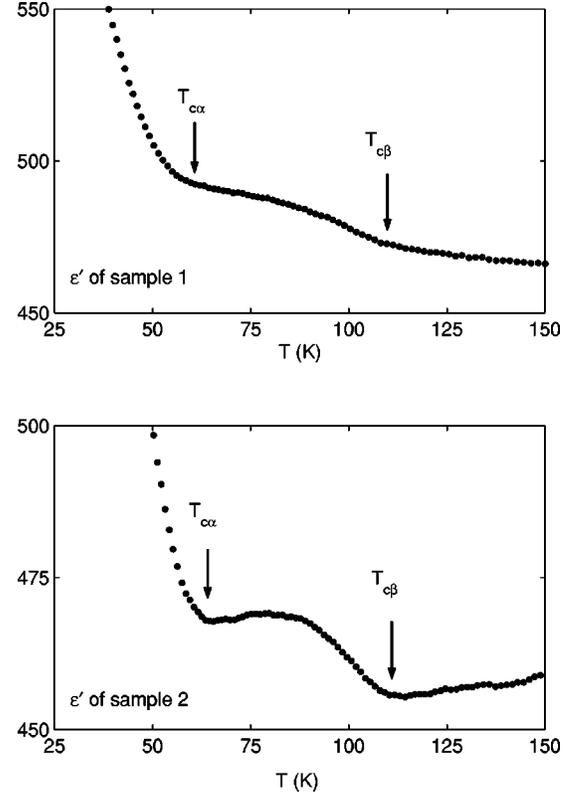


FIG. 2. Dielectric constants  $\epsilon'$  at 10 GHz of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  for samples 1 and 2. Note the onset of dielectric modes at  $T_{c\alpha}=60$  K and  $T_{c\beta}=110$  K are present in both the samples.

all temperatures. We believe  $\tilde{\epsilon}_\gamma$  is the contribution which has been measured by several previous investigators<sup>2,3</sup> on nonmetallic  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  and represents a polarization mode formed at high temperatures  $T > 300$  K.  $\tilde{\epsilon}_\alpha$  and  $\tilde{\epsilon}_\beta$  indicate the onset of two dielectric modes which turn on below transition temperatures  $T_{c\alpha}=60$  K and  $T_{c\beta}=110$  K. We describe these modes with the following parameters:

$$\epsilon_{\beta 0}(T) = 60[1 - (T/T_{c\beta})], \quad T_{c\beta} = 110 \text{ K} \quad \text{and} \quad \tau_\beta(T) = 4 \times 10^{-10} (\text{sec} \cdot \text{K}) / (T + 200), \quad \text{for the } \tilde{\epsilon}_\beta \text{ process, and}$$

$$\epsilon_{\alpha 0}(T) = 280[1 - (T/T_{c\alpha})], \quad T_{c\alpha} = 60 \text{ K} \quad \text{and} \quad \tau_\alpha(T) = 2.5 \times 10^{-10} (\text{sec} \cdot \text{K}) / (T + 5), \quad \text{for the } \tilde{\epsilon}_\alpha \text{ process.}$$

$\epsilon_{\alpha 0}$  and  $\epsilon_{\beta 0}$  are similar to order parameters which grow at temperatures below a transition. As  $T$  is lowered, both  $\epsilon'_\alpha(T)$  and  $\epsilon''_\alpha(T)$  increase initially due to the growing polarization. However below a characteristic temperature both  $\epsilon'_\alpha(T)$  and  $\epsilon''_\alpha(T)$  begin to decrease because the dipoles are no longer able to follow the microwave field. The peak temperature  $T_{p\alpha} \sim 25$  K is determined by the condition  $\omega\tau_\alpha(T_{p\alpha}) = 1$ , although the peak for  $\epsilon'$  is at a higher  $T$  than for  $\epsilon''$ . The peaks are so-called dielectric loss peaks. Identical arguments hold for  $\tilde{\epsilon}_\beta(T)$  also. Here the peak is much broader and occurs at  $T_{p\beta} \sim 75$  K.

For the  $\alpha$  and  $\beta$  processes, the temperature dependence is too broad to be described by an activated relaxation rate. We have found that a relaxation rate which is linear in  $T$ , i.e.,  $\tau_{\alpha,\beta}^{-1}(T) \approx a_{\alpha,\beta}(T + T_{(\alpha,\beta)0})$ , describes the data very well as seen in Fig. 1, with  $a_\alpha \approx 0.4 \times 10^{10} (\text{sec} \cdot \text{K})^{-1}$ ,  $T_{\alpha 0} = 5$  K

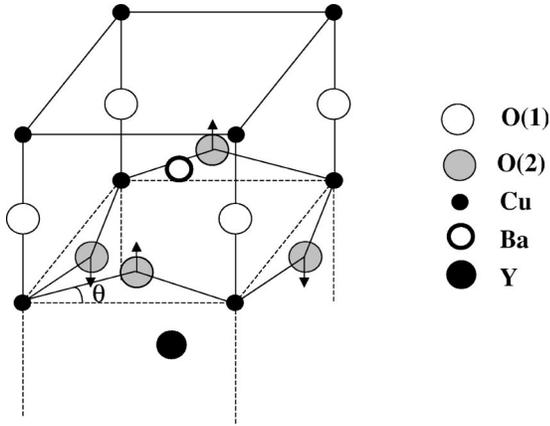


FIG. 3. Apical, O(1) and planar O(2) oxygens in YBCO. The change in the buckling angle at 110 K changes the polarizability of O(1) resulting in the  $\beta$  dielectric mode.

and  $a_\beta \approx 0.25 \times 10^{10} (\text{sec} \cdot \text{K})^{-1}$  and  $T_{\beta 0} = 200$  K. Such relaxation rates with linear  $T$  dependences are well known in the copper oxide superconductors.

We note that similar large dielectric constants have been observed in other copper oxides. In  $\text{Bi}_2\text{Sr}_2(\text{Dy}, \text{Y}, \text{Er})\text{Cu}_2\text{O}_8$ , the parent compound of Bi:2212, large in plane  $\epsilon' \sim 10^3 - 10^5$  were reported<sup>11</sup>. It is also important to note that the dielectric modes discussed here bear a strong similarity to the numerous modes observed in the dielectric response of the perovskite  $\text{SrTiO}_3$  (Ref. 12) at 65, 37, and 16 K. The dielectric loss peaks reported here are similar to those observed in  $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$  (Ref. 13) and  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ .<sup>14</sup>

The present results indicate that at 110 (Ref. 15) and 60 K, two polarization onset transitions occur in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ . We note that the 110 and 60 K onsets cannot arise from any contamination of the sample by a superconducting phase, since then the contribution should be diamagnetic (negative  $\epsilon'$ ), opposite to what is observed.

A scenario leading to such dielectric transitions can be arrived at starting with the so-called Bilz model for ferroelectricity,<sup>16</sup> which is based upon the nonlinear polarizability of oxygen, and originally developed for perovskite structures. This applies to a displacive-type ferroelectric where dipole moments are induced during the phase transition so that soft mode concept becomes important. Above the displacive ferroelectric transition, the oxygen atoms are essentially oscillating in a potential well. Below the ferroelectric transition, a double-well is formed and the oxygen atom then locks into one of the minima—the displacement then leads to a large permanent polarization. In the present case the ferroelectricity is prevented from occurring, either due to quantum fluctuations, as was proposed for  $\text{SrTiO}_3$ ,<sup>17</sup> or due to coupling between Ba–O and Cu–O layers (Fig. 3).<sup>6</sup> Consequently the O potential is greatly softened leading to the large dielectric permittivities observed.

We use a modification of the Bilz model specifically for the oxo-cuprate superconductors implemented by Shenoy, *et al.*<sup>6</sup> The equation-of-motion of the oxygen relative ion-electron coordinate  $\vec{w}$  is given by  $m_e(\ddot{\vec{w}} + \Gamma\dot{\vec{w}}) + D\vec{w} = Ze\vec{E}e^{-i\omega t}$ , where  $D$  is the self-consistent harmonic ap-

proximation (SCHA) curvature of the anharmonic Oxygen electronic potential. For a driving electric field  $\vec{E}e^{-i\omega t}$ , the susceptibility  $\alpha = Ze w/E = Z^2 e^2/m_e(\omega_0^2 - \omega^2 - i\omega\Gamma)$ . The dielectric constant  $\epsilon = n\alpha/\epsilon_0$  then becomes

$$\tilde{\epsilon} = \frac{\epsilon(0)}{(1 - \omega^2/\omega_0^2) - i\omega\tau}. \quad (3)$$

Here  $\epsilon(0) = nZ^2 e^2/D$  and  $\tau = \Gamma/\omega_0^2$ .

Rather large dielectric constants are feasible for soft modes. For the case of the O atom in the Ba–O layer in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  we have  $n = 1.2 \times 10^{28} \text{ m}^{-3}$ . With  $\epsilon_0 = 8.85 \times 10^{-12} \text{ F/m}$ , and using the resonance frequency,  $f_0 = 3 \times 10^{13} \text{ Hz}$  ( $\omega_0 = 2\pi f_0$ )<sup>18</sup> so that  $D (= Zm_e\omega_0^2) = 3.3 \times 10^{-2} \text{ N/m}$ , we get  $\epsilon(0) \sim 10^3$  comparable to the experimental results. Note that despite the softening, the condition  $\omega \ll \omega_0$  is well satisfied ( $f = 10^{10} \text{ Hz}$ ), and the so-called Drude-Lorentz form of a resonant mode given by Eq. (3) reduces to the Debye-like relaxation forms used in Eq. (2). The above estimate indicates considerable softening of the anharmonic O potential. Indeed this can happen because the curvature is extremely sensitive to interatomic forces in these materials. A mechanism for such softening has been given by Shenoy *et al.*<sup>6</sup> for the layered HTS.

There have been extensive studies of lattice dynamics in superconducting oxides, especially in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ .<sup>19</sup> One of the key features that has emerged is that there are small structural distortions which occur although there is no change in the overall structure. Particularly well-established are the structural distortions reported at  $T_c$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ ,  $\text{Hg}_1\text{Ba}_2\text{CuO}_4$ , and  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ .<sup>20</sup> Also, lattice instabilities have been reported, in the dilatometry measurements on doped and undoped  $\text{La}_2\text{CuO}_{4+x}$ , at 32 and 36 K that are found to be independent of the density of charge carriers.<sup>21</sup> In  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  the coupling between apical O(1) and planar O(2) oxygen changes at the superconducting transition due to the displacements of O(2) in the directions perpendicular to the CuO<sub>2</sub> plane (Fig. 3) changing the buckling in the plane.<sup>19</sup> The nearness of the  $\beta$  dielectric mode around 110 K to the  $T_c = 93$  K ( $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ ) is striking and suggests a possible change in the O(2) dynamics in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  (YBCO) as well, and hence we attribute this  $\beta$  mode to the change in the dynamics of O(2).

In Ref. 6 the possibility of dielectric modes in coupled Ba–O and Cu–O layers is described. Including the various interatomic forces, it can be shown that the curvature  $D$  becomes a function of the buckling angle  $\theta$ . The change in buckling at 110 K would lead to a change in the mixing of the  $ab$  plane acoustic and  $c$ -axis optic mode, resulting in a new set of mixed modes which would move to lower frequency due to softening. The resulting decrease in  $D$  would then explain the  $\beta$  mode at 110 K. The calculation of Shenoy *et al.*<sup>6</sup> based on the mean-field approach support our description of the dynamics of O(1).

The presence of the 60 K mode suggests another local structural change at this temperature scale, possibly in the chain layer. It is very interesting to note that this temperature scale is present in the doped YBCO as well.<sup>25</sup> In addition to

the proximity of the 110 K transition to the optimum superconducting  $T_c$  of 93 K noted above, equally important is the presence of a secondary temperature scale around 60 K in certain measurements of  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ .<sup>10,22–24</sup> This temperature scale has manifested itself in various experiments whose connections are becoming apparent only recently. Thus the present results may have important implications for superconductivity in these materials.

In optimally doped single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  an additional onset of pair conductivity at 65 K was noted in Ref. 10, well below the main  $T_c=93$  K. The consequences of this on the thermal conductivity<sup>22</sup> and vortex transport<sup>23</sup> have been observed. The present temperature scales also have striking resemblance to transitions around 110 and 65 K in over doped YBCO observed in nuclear quadrupole resonance (NQR) measurements by Grevin *et al.*,<sup>24</sup> which have been interpreted in terms of charge-density wave (CDW) correlations. In their results a short-range CDW sets in the Cu–O chains at 110 K which becomes long range around 65 K. The formation of CDW in the chains modulate the charge in the planes leading to a transition between an inhomogeneous charge state to a low-temperature ordered charge state in the planes. Keeping in view the fact that NQR probes the electric field gradient around the Cu nucleus the NQR transitions could as well be due to changes in local structure which leads to CDW formation in the doped metallic YBCO. Together, the NQR, thermal conductivity and the present results stress the importance of the 110 and 60 K temperature

scales. These results indicate that local distortions in the structure at 110 and 60 K would lead to changes in polarization as we have observed in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  and charge ordering in doped YBCO. Microwave conductivity measurements on doped crystals<sup>25</sup> from  $x=0$  to 1, when taken together with these other observations, strongly suggest that doping moves this onset temperature from 60 K at  $x=0$  to  $\sim 70$  K at  $x=1$ . Also the superconducting  $T_c$  increases with  $x$ , but never exceeds 93 K. An interesting implication is that the implied structural distortion transition at 110 K may represent an upper limit on the superconducting transition,  $T_c$ .

In conclusion we have observed three paraelectric modes with temperature onsets at 60, 110, and  $>300$  K in the non-metallic  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ . These modes are well described by the change in polarizability of apical O(1) oxygens due to change in lattice dynamics with temperature. A striking observation is that the two low-temperature modes have direct connections with the transitions observed by traditional lattice probes as well as NQR and thermal conductivity measurements on doped superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0+x}$ , strongly suggesting that oxygen and lattice dynamics play an important role in both the superconducting and nonsuperconducting materials.

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