Different regions of fluctuation conductivity in unirradiated and alpha-irradiated $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $(Bi,Pb)_2Sr_2CaCu_2O_{8+\delta}$ superconductors

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Abstract

Radiation damage in Bi-2212 and $(Bi_{0.92}Pb_{0.17})$-2212 superconducting pellets due to irradiation by 40 MeV He^{++}-beam has been studied, as these two compounds differ with respect to lattice strain in their Bi–O and (Bi,Pb)–O layers. These samples were prepared with excess oxygen that accounts for their lowered superconducting transition temperature, T_c, of 59.5 and 64.5 K, respectively. Irradiation to 8.14×10^{15} He^{++}/cm^2 raised T_c by 4.5 and 5.5 K, respectively, and also their normal state resistivities. Excess conductivity analysis for the unirradiated and irradiated samples showed (i) a transition of the conductivity fluctuation from 2D ($\lambda = 1.0$) to 3D ($\lambda = 0.5$) behaviour, on cooling across a temperature T_0, and (ii) also the signature ($\lambda = 3.0$) of critical fluctuations in a network of superconducting microregions, for temperatures below T_{cr}, where T_{cr} is seen to be 5 to 10 K lower than T_0. The rise in T_c has been explained from irradiation-induced removal of oxygen taking the hole concentration towards the value for the peak in T_c.

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1. Introduction

The widely undertaken analysis of excess electrical conductivity, $\Delta\sigma$, in high temperature superconductors (HTSC) from different [1–3] fluctuation theories has concentrated mostly on investigating a crossover [4–6] from a 3D fluctuation (exponent, λ, approaching 0.5) at low temperatures to a 2D one (λ approaching 1.0) on warming across a crossover temperature, T_0, and on the inter-layer coupling constant J [7–9]. In terms of the reduced temperature $t = (T - T_c^{mf})/T_c^{mf}$, where T_c^{mf} is the mean field superconducting transition temperature, Aslamazov–Larkin [1] equation has the form: $\Delta\sigma(t) = At^{-\lambda}$ with A denoting a temperature-independent constant. On the one hand, such analysis has been interesting on account of the enhancement of the fluctuations for various HTSC due to their layered quasi-2D structure and small coherence length. On the other hand, it has often been difficult to choose the temperature range for fitting due to various

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possible contributions to the variation of $\Delta\sigma$ with temperature, T. The present paper has addressed this problem.

In the present work, a region with the exponent [10,11] approaching 3.0 and implying a critical fluctuation has been identified below the temperature T_{cr}, where T_{cr} is much lower than T_0. It is shown that any attempt to extend the fit with exponents approaching 0.5 into this critical region will only add error and ambiguity to the results.

Jurelo et al. [10,11] observed these regions in differently heat treated pellets of $(Bi,Pb)_2Sr_2CaCu_2O_{8+\delta}$ or (Bi,Pb)-2212 and (Bi,Pb)-2223. Their heat treatments provided samples with different disorders or different oxygen contents as characterized by their superconducting critical temperature T_p, varying from 64.4 to 78.4 K for (Bi,Pb)-2212. Here, 40 MeV He^{++}-irradiation has been utilized to knock off some of the excess oxygen from our furnace cooled Bi-2212 and $(Bi_{0.92}Pb_{0.17})$-2212 or (Bi,Pb)-2212 pellets. So the second objective of this work is to gain a deeper insight into the radiation damage through our detailed analysis of excess conductivity. The interest in the furnace cooled Bi-2212, as one of our starting samples is its apparently opposite response to radiation damage, as elaborated in the next paragraph. The compound (Bi,Pb)-2212 has been chosen [7–9] as a bridge [12–14] between Bi-2212 and (Bi,Pb)-2223.

Radiation damage introduces defects in a solid. So it generally reduces the superconducting critical temperature, T_c. But we earlier observed [4–9] an alpha-irradiation-induced increase in T_c in a Bi-2212 pellet, that was furnace-cooled to have an excess oxygen and hence a lowered T_c. The irradiation knocking off some of the excess O-atoms from the lattice sites has been identified as the cause [4–6,15] of the observed rise in T_c. In this work, the direction and magnitude of change in T_c for (Bi,Pb)-2212 due to 40 MeV He^{++}-irradiation will be probed and the rise confirmed for Bi-2212 by irradiating these two samples simultaneously. These (Bi,Pb)- and Bi-samples were produced simultaneously by the same furnace cooling, there being no other damage studies on (Bi,Pb)-2212. However, the partial substitution of Bi by larger Pb ions in (Bi,Pb)-2212 tends to expand the Bi–O spacings and makes it interestingly different from that in Bi-2212 with respect to binding of the O-atoms, that are most affected by such He^{++}-irradiation.

2. Experimental outline and data analysis

Polycrystalline superconductors Bi-2212 and $(Bi_{0.92}Pb_{0.17})$-2212 were prepared by solid state reaction [7–9,12–14]. Starting with the oxides or carbonates in such proportions as to give the desired cation composition, the samples were prepared by repeated grinding, pelletizing and firing till the X-ray diffraction studies (using Philips PW 1710 diffractometer) revealed peaks for the desired phase only. Samples were finally cooled slowly to incorporate excess oxygen. Critical temperature, T_c, of the as prepared Bi-2212 and (Bi,Pb)-2212 samples were determined from four probe resistivity measurement in a Leybold Cryogenerator set-up complete with a Keithley 182 digital nanovoltmeter and a Lakeshore 120 current source.

The samples were simultaneously irradiated by the same 40 MeV α-particles, obtained from the Variable Energy Cyclotron Centre in Calcutta. The range of 40 MeV α-particles in Bi-2212 and (Bi,Pb)-2212 samples is about 250 μm. The samples were made thinner to ensure that the α-particles passed out of the samples, causing radiation damage and no implantation. During irradiation, samples were placed, with good thermal contact, on an aluminium target holder [7–9] of considerable thermal mass. By earthing, the insulated target holder through a current integrator, the beam current and, hence, the fluence were accurately measured. The two samples were irradiated together by the 40 MeV α-beam. The second half of each dose was allowed on the samples after turning the samples by 180°. The damage profiles for irradiation from two sides add up to give a more uniform damage throughout the thickness than is possible by irradiation from one side. The irradiated samples were examined again by XRD and their T_c values determined by the four probe resistive method.

We have analyzed our resistivity vs. temperature experimental data by adopting the simplest fluctuation conductivity approach [10,11]. The fluctuation induced enhancement in conductivity, termed as excess conductivity or para-conductivity, $\Delta\sigma$, can be described by the relation

$$\Delta\sigma = A_f(T - T_f)^{-\lambda}. \tag{1}$$

Here, λ is the critical exponent, A_f and T_f are fitting constants and $\Delta\sigma = \sigma_m - \sigma_n = 1/\rho_m - 1/\rho_n$, where the subscripts m and n refer, respectively, to the measured and normal values of conductivity σ and resistivity ρ. The normal resistivity ρ_n, at low temperature, was estimated by extrapolating the high temperature behaviour $\rho_n(T) = a + bT$. The constants a and b in the above equation have been determined by a least square fitting of our ρ vs. T data in the higher temperature part of the ρ vs. T graph, which is linear.

From Eq. (1), we get

$$\frac{\lambda}{(T - T_f)} = -\frac{d}{dT}[\ln(\Delta\sigma)] = \chi_\sigma. \tag{2}$$

A plot of χ_σ^{-1} (calculated from the observed T-dependence of $\Delta\sigma$) vs. T for data in various regions allowed the determination of various pairs of λ and T_f.

3. Results and discussion

The XRD patterns of the unirradiated (Bi,Pb)-2212 sample, (Fig. 1(a)) and the unirradiated Bi-2212 sample show peaks for the 2212 phase only. The patterns, taken after He^{++}-irradiation, show that for both samples, there is a gradual growth of an amorphous phase with irradiation dose. The patterns for irradiated (Bi,Pb)-2212 are shown in Fig. 1(b) and (c).

The same furnace cooling resulted (Fig. 2) in $T_c(R=0)$ of 64.5 K for Bi-2212 and 59.5 K for (Bi,Pb)-2212. Since T_c depends [15] on the O-content (controlling the hole type carrier concentration) according to a bell-shaped curve, the presently observed lower T_c must have resulted from O-content either higher or less than the optimum giving the maximum T_c. Annealing the samples in argon or preparation by quenching instead of slow furnace cooling resulted [12–14] in higher T_c proving the present samples to have

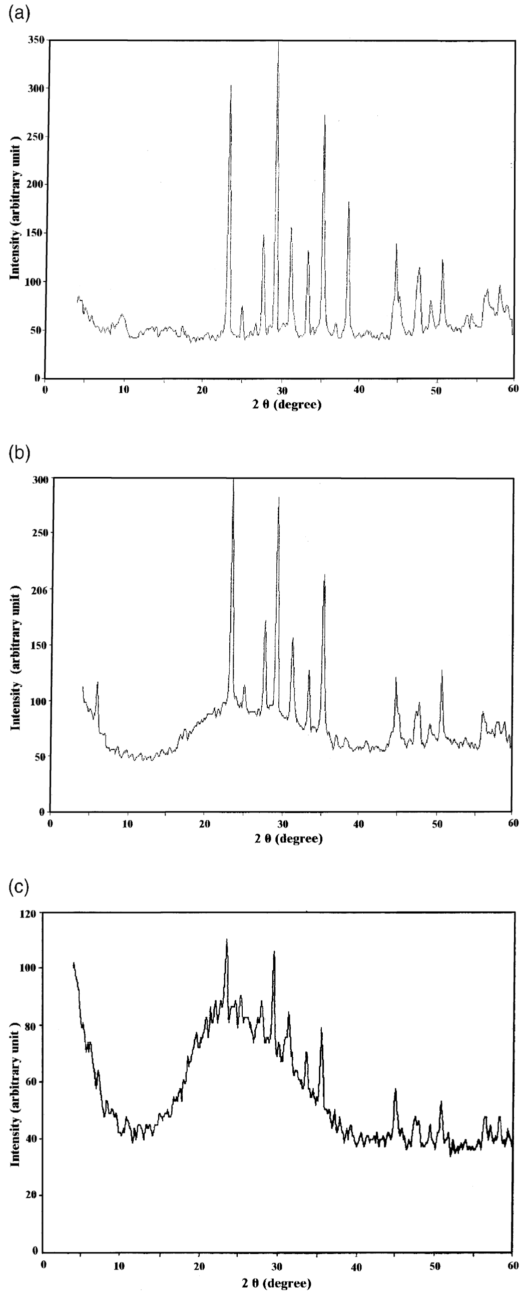
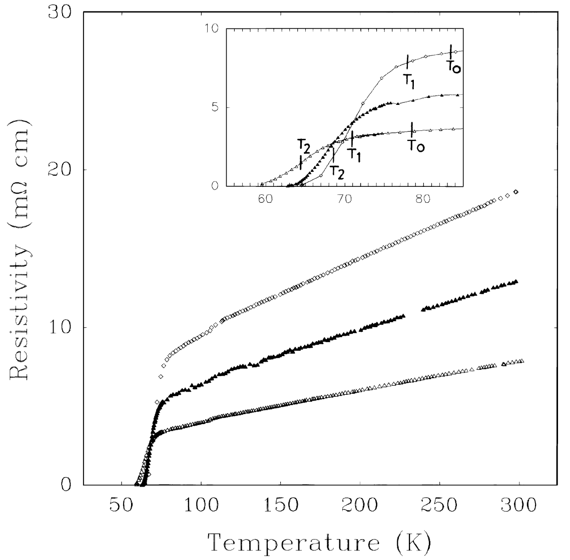


Fig. 1. X-ray powder diffraction pattern (for CuK_α) for different (Bi,Pb)-2212 samples : (a) unirradiated, (b) irradiated with 2.2×10^{15} He^{++}/cm^2, and (c) irradiated with 8.14×10^{15} He^{++}/cm^2.

higher or excess oxygen. Since the maximum possible T_c is essentially the same in Bi-2212 and

(a)



(b)

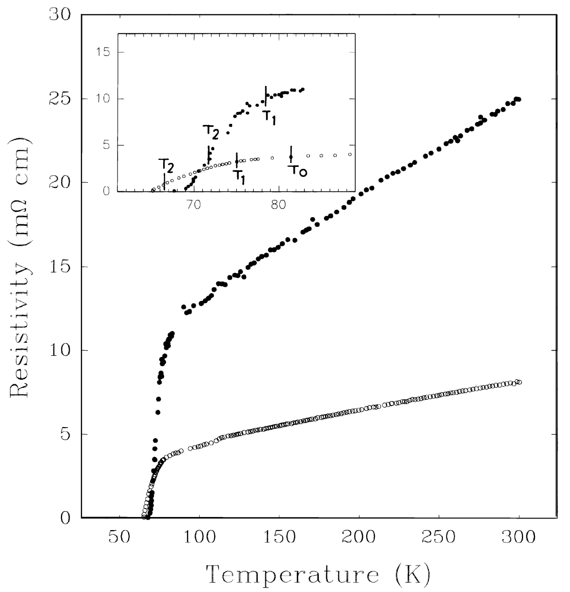


Fig. 2. Electrical resistivity as a function of temperature for (a) (Bi,Pb)-2212, for samples He^{++}-irradiated to 8.14×10^{15} cm^{-2} (□) and to 2.2×10^{15} cm^{-2} (▲) and the unirradiated sample (△), and (b) Bi2212 for He^{++} irradiated to 8.14×10^{15} cm^{-2} sample (○) and unirradiated sample (●). Insets (expanded views of the transition regions) and Table 1 give T_0, $T_1 = T_{cr}$ and $T_2 = T_p$.

(Bi,Pb)-2212, the difference in the unirradiated T_c for our SL-C Bi- and (Bi,Pb)-samples can be due either to different graphs for the two compounds or to more O-intake by (Bi,Pb)-2212.

Our α-irradiation shifts the resistivity vs. temperature plot similarly for the two compounds, increasing $T_c(R=0)$ and the normal state resistivity in both cases. In fact, 8.14×10^{15} He^{++}/cm^2 increases $T_c(R=0)$ by 5.5 K for (Bi,Pb)-2212, and by 4.5 K for Bi-2212. Fig. 2 depicts, the results obtained (a) for (Bi,Pb)-2212, the new system that has never been studied under particle irradiation, and (b) for Bi-2212. For (Bi,Pb)-2212, an additional irradiation to an intermediate fluence of 2.20×10^{15} He^{++}/cm^2 has been seen to result in a graph lying between the graphs for unirradiated (U) and 8.14×10^{15} He^{++}/cm^2-irradiated (I) samples. These figures confirm that He^{++}-irradiation can increase T_c if it is less than the maximum possible on account of containing excess oxygen.

Jurelo et al. [10,11] have explained [16,17] that the newly observed region with Aslamazov–Larkin exponent approaching 3.0 arises from the critical behaviour of a network of superconducting "particles" dispersed in a less conducting matrix, the classical example being the packing of metallic superconducting spheres or particles. They attribute the temperature region immediately below T_{cr}, shown as T_1 in our Fig. 2(a), to a network of microscopic "grains" of HTSC within individual HTSC grains or crystallites. This so-called microscopic intragrain granularity is due to the smallness of the coherence length and the consequent coupling of microscopic defects (e.g. deviations from stoichiometry) and spatial fluctuation of T_c. Here one must cite our earlier SEM and EDAX observations [18] of spatial variation of large magnitude in cation composition, in addition to the well-known O-inhomogeneity. Such non-stoichiometry, reported also by others, points to abovementioned microscopic defects. At still lower temperatures, lower than the bulk transition temperature, $T_p = T_c^{mf}$, and approaching $T_c(R=0)$, Jurelo et al. have identified [10,11,16,17] fluctuation conductivity diverging with exponents approaching 3.0 (for the mesoscopic granularity in the network of crystallites or grains measuring 0.1–20 μm) and 4.0 (a vortex-glass exponent, also

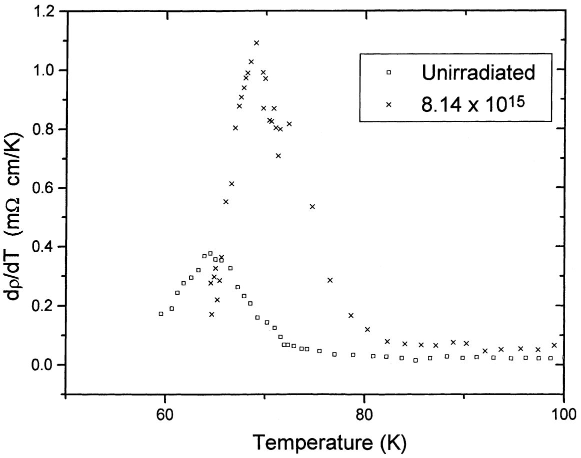


Fig. 3. The variation of the temperature derivative of the electrical resistivity as a function of temperature for unirradiated (□) and irradiated (×) (Bi,Pb)-2212 samples. Irradiation fluence in He^{++}/cm^2 has been indicated in the figure.

related to granularity). The supposedly vortex-glass exponent [10,11] has been observed only at higher current densities, not used by us. The 3D Gaussian fluctuation (above T_{cr}) and microscopic critical fluctuation (below T_{cr}) have been seen to depend on the nature of the sample.

Each of our samples shows a sharp peak in $d\rho/dT$ and possibly a narrow shoulder on the high temperature side of the peak, before as well as after the radiation damage. This is shown for (Bi,Pb)-2212 in Fig. 3. Data of Jurelo et al. [10,11] in their Fig. 1(b) appear to show such shoulders below and above the peak for Bi-2212. Pureur et al. [10,11] report a clear shoulder below the peak, for Y-123, relating the shoulder to a thermally controlled percolation-type phenomenon and mesoscopic and microscopic inhomogeneities. The position of the peak, called T_p by Pureus et al., [10,11] actually gives the mean field superconducting transition temperature, T_c^{mf}. So T_c^{mf} is seen to shift (Table 1 and Fig. 2) towards higher temperature by irradiations, just like $T_c(R=0)$. The sharp peak implies only one superconducting phase in the sample.

Fig. 4 is a plot of χ_σ^{-1} (defined earlier) vs. T for (Bi,Pb)-2212. The corresponding plot for Bi-2212 has been similar. The critical fluctuation due to microscopic granularity [10,11] is clearly proved from our fit with the exponent (λ(cr)) of 3.17 (for the unirradiated (Bi,Pb)-2212) or 3.27 (for the irradiated (Bi,Pb)-2212) at temperatures lower than $T_{cr} = 70.9$ or 77.9 K. The shortened notation T_1 has been used to mark T_{cr} in Fig. 2. Similarly T_c^{mf} or T_p has been indicated in the figures as T_2. These temperatures indicate transitions between different regions and mark the regions of fitting. On the highest temperature side, the fitting range is terminated on reaching the linear ρ vs. T-region. At temperatures below about 64 K, the data points for the unirradiated sample appear to fit a different graph (not drawn) with roughly the same slope (i.e. same exponent) but shifted upwards, that should represent [10,11] critical fluctuation in mesoscopic grains.

The fit in the temperature range higher than T_{cr} but below T_0 (Table 1) corresponds to λ(3D) nearly equal to 0.5, that represents 3D Gaussian fluctuation. The critical fluctuation and 3D fluctuation graphs intersect at T_{cr}. 2D fluctuation regions can be concluded for the irradiated and unirradiated samples from the exponents being 0.97 and 0.91, respectively, for temperatures above T_0. These 2D fluctuation graphs intersect the corresponding 3D fluctuation graphs at $T_0 = 83.5$ K for the irradiated and at $T_0 = 78.4$ K for the unirradiated (Bi,Pb)-2212 sample. This better

Table 1
Consolidated result of our analysis of fluctuation conductivity in 2D, 3D and critical (cr) regimes of temperature for unirradiated (U) and 8.14×10^{15} He^{++}/cm^2-irradiated (I) (Bi,Pb)-2212 and Bi-2212 oxide superconductors

Sample	T_c (K)	λ (2D)	T_0 (K)	λ (3D)	T_{cr} (K)	λ (cr)	T_c^{mf} (K)
U (Bi,Pb)-2212	59.5	0.91	78.4	0.59	70.9	3.17	64.22
I (Bi,Pb)-2212	65.0	0.97	83.5	0.48	77.9	3.27	68.6
U Bi2212	64.5	1.10	81.4	0.53	75.0	3.05	66.4
I Bi2212	69.0	0.97	87.1	0.55	78.2	2.90	71.7

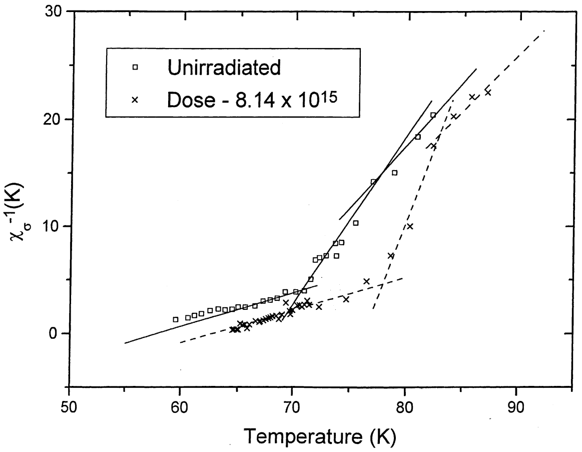


Fig. 4. The variation of $\chi_\sigma^{-1} = -[\mathrm{d}[\ln(\Delta\sigma)]/\mathrm{d}T]^{-1}$ vs. temperature, T, where $\Delta\sigma$ represents the excess conductivity for unirradiated (□) and irradiated (×) (Bi,Pb)-2212 samples. Irradiation fluence in He^{++}/cm^2 has been indicated in the figure.

known 3D to 2D crossover on warming across T_0 was very clearly seen in our earlier work [4–9] with more data points above T_0. Table 1 presents the results for (Bi,Pb)-2212 and Bi-2212 samples.

Here it should be recalled that, by microscopic granularity, authors [10,11] imply superconducting regions within the same grain that are microscopically different from each other due to intragranular variation of local T_c or due to intragranular defects like composition variation [10,11,18]. Interaction among grains, the so-called mesoscopic granularity, has showed up only for the unirradiated samples, for unirradiated (Bi,Pb)-2212 below about 64 K (Fig. 4). This mesoscopic region [10,11] (lowest temperature data points on an upwardly shifted parallel graph) is absent for the irradiated sample in Fig. 4. It is most probable that the enhancement of intragranular defects by irradiation has made the microscopic (intragranular) and mesoscopic (intergranular) fluctuation regions indistinguishable in the irradiated sample. This fine internal fragmentation of the sample due to irradiation is proved also from the gradual but dramatic growth of the amorphous phase in the irradiated samples (Fig. 1(b) and (c)). The present work has concentrated on and identified critical fluctuation regions for unirradiated and irradiated Bi/(Bi,Pb)-2212 superconductors.

4. Conclusions

The superconducting critical temperature has increased, due to 40 MeV He^{++}-irradiation, by 5.5 K in $(Bi_{0.92}Pb_{0.17})$-2212 or (Bi,Pb)-2212 pellets with unirradiated $T_c(R=0)$ of 59.5 K and by 4.5 K in Bi-2212 with unirradiated $T_c(R=0)$ of 64.5 K. Loading excess oxygen by the same furnace cooling had resulted in these samples. Quenching the same pellets increase $T_c(R=0)$ towards 80 K, as quenching as well as the above irradiations remove lattice oxygen atoms. This shifts the carrier concentration towards the value for the peak in T_c.

Using a new technique based on logarithmic temperature derivative of the excess conductivity, critical fluctuation regions appropriate to granular superconductors and identified by the Aslamazov–Larkin exponent approaching 3.0 have been observed just above the transition to zero electrical resistance. This recognizes the realities of granularity and microgranularity in high T_c superconductors. Here, depending mainly on the temperature regime, the HTSC grains (or crystallites) of the pellet and the intracrystallite micro-granular regions play the role of metallic superconducting particles [16,17] artificially dispersed in a less conducting matrix, the classical example, where the exponent approaches 3.0. Here an effort has been made in irradiated and unirradiated HTSC samples to view the critical fluctuation regions (exponent = 3.0) along with the better investigated regions with exponents approaching 0.5 and 1.0 that imply 3D and 2D fluctuations.

It is further noted that mesoscopic and microscopic granularities can be distinguished only for the unirradiated samples. Generation of intragranular defects by the present irradiation has been sufficient to make the two kinds of granularities indistinguishable in the low temperature fluctuation data. This radiation damage shows up also in the X-ray diffraction patterns.

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