



Article

The Electron–Phonon Interaction in Non-Stoichiometric $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ Superconductor obtained from the Diffuse Elastic Scattering of Helium Atoms

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Abstract: Previously, helium atom scattering (HAS) has been shown to probe the electron–phonon interaction at conducting crystal surfaces via the temperature dependence of the specular peak intensity. This method is now extended to non-stoichiometric superconductors. The electron–phonon interaction, as expressed by the mass-enhancement factor λ , is derived from the temperature dependence of the diffuse elastic scattering intensity, which specifically depends on the non-stoichiometric component responsible for superconductivity. The measured value of the mass-enhancement factor for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ at the optimal doping $\delta = 0.16$ is $\lambda = 0.55 \pm 0.08$ in good agreement with values of λ recently estimated with other methods. This also confirms the relevant role of electron–phonon interaction in high-temperature non-stoichiometric cuprate superconductors.

Keywords: superconductivity; non-stoichiometric cuprates; HAS scattering



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1. Introduction

The layered compound $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212) owes its hole-doping and high-temperature superconductivity to a deviation from stoichiometry, spontaneously produced in the growth process [1] and consisting of an excess δ of oxygen ions, essentially concentrated in the Bi–O layers. Superconductivity is observed for roughly $0.09 < \delta < 0.29$, with a maximum $T_c = 95$ K at about $\delta = 0.17$ [2]. Doping with additional oxygen atoms, besides injecting free holes and producing static deformations of the stoichiometric lattice, also affects the electron and phonon structure by introducing localized and resonance states, which may eventually play a role in the superconducting properties activated by the doping itself [3]. A natural question is whether the electron–phonon (EP) interaction is also significantly affected by doping. High- T_c superconductors, notably Bi-2212, do not explicitly rely on the EP interaction like traditional BCS superconductors, although the EP interaction has been shown to largely contribute to its superconductivity, with a mass-enhancement factor $\lambda \approx 0.6$ as the optimal doping level [4,5]. Recently, O’Mahony et al. [6] have provided experimental evidence in favor of Anderson superexchange [7] as the dominant pairing mechanism in high- T_c superconductivity in Bi-2212. Another possible mechanism for high- T_c superconductivity is doped cuprates, formulated by Hiroyasu Koizumi [8], and is based on the formation of small polarons and associated spin vortices in the bulk CuO_2 layers allowing for a d-wave pairing. Their spin-moment texture has been

predicted to be accessible to surface probes like angle-resolved photoemission spectroscopy (ARPES) and scanning tunnelling spectroscopy (STS) [9].

In the last decade it has been demonstrated that He atom scattering (HAS) from conducting crystal surfaces permits a direct determination of the EP interaction, as represented by the mass-enhancement factor λ , from measurements of the temperature dependence of the Debye–Waller (DW) exponent, as derived from the elastic specular or diffraction peak intensities [10,11]. This is based on the fact that at conducting surfaces, He atoms are repelled by the surface electron density and can excite (annihilate) phonons via the phonon-induced modulation of the surface-electron density, i.e., via the EP interaction. Thus, HAS can probe the atomic motions deep beneath the surface at the range of EP interaction (*quantum sonar effect* [10]).

In inelastic HAS time-of-flight (TOF) spectra, currently used to measure the dispersion curves of surface phonons [12], there is always an additional peak at zero-energy transfer, called the diffuse elastic (DE) peak. This peak originates from the elastic scattering of surface defects, or, in general, from any feature which perturbs the surface periodicity. Thus, the contribution from static and dynamic effects of doping to the EP interaction in doped superconductors such as Bi-2212 can be extracted from the temperature dependence of the HAS DE peak intensities.

Here, we report on HAS TOF measurements from Bi-2212 at optimal doping. These provide, besides a set of surface phonon dispersion curves in the range of energy below 10 meV [13], the DE peak intensities at two different temperatures, 100 and 310 K. The EP interaction $\lambda = 0.55 \pm 0.08$ derived for Bi-2212 from the two different temperatures at about the optimal doping condition is in good agreement with what is currently known from other methods [4,5,14]. This supports the present conjecture that the DE peak can also be used to extract information on the EP interaction at conductive surfaces where the conductivity is essentially induced by doping.

It is important to note that in the specific case of high- T_c cuprates, where the unit cell includes several atomic layers and superconductivity is essentially associated with copper oxide layers, the critical temperature does not decrease in ultrathin films, even down to a single-cell thickness corresponding to one formula [15]. In this regard, bulk $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ behaves as a genuine 2D superconductor, which is equivalent to saying that HAS-DE data are providing in this case information on the actual λ of the bulk material.

2. He Atom Scattering Time-of-Flight Data

In previous HAS diffraction experiments [13], the structure of the Bi-O(001) surface of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, with $\delta = 0.16$ at about the optimal doping condition and corresponding $T_c = 91$ K, revealed, at temperatures well above T_c , an incommensurate striped superstructure in the $\langle 1\bar{1}0 \rangle$ direction with a rectangular supercell approximated to $(\sqrt{2}a, 5\sqrt{2}a)R45^\circ$, as compared to the ideal square unit cell of edges (a, a) , where $a = 3.814$ Å is the Bi-O distance [13]. This superstructure, detected by HAS diffraction in the reciprocal space, was later clearly imaged by Zeljkovic et al. [16] using scanning tunnelling microscopy (STM) at increasing critical temperatures up to the maximum $T_c \sim 91$ K. The analysis by Zeljkovic et al. at the optimal oxygen doping level of $\sim 4\%$ per CuO_2 reveals random interstitials in the Bi-O plane, and similarly other interstitials at $\sim 2.5\%$ per CuO_2 in the Sr-O plane (type-B and type-A oxygen, respectively [17]). The association of superconducting properties to the inhomogeneities produced by non-stoichiometric oxygen also supports the present derivation of the EP interaction from the measured temperature dependence of the HAS DE peak.

Figure 1 displays the TOF spectra in the planar 90° scattering-angle configuration at four different incident angles ($\theta_i = 40^\circ$ to 43°) for the same incident He atom momentum $k_i = 5.8$ Å $^{-1}$ (incident energy $E_i = 18$ meV) and for surface temperatures of 100 K (panel (a)) and 310 K (panel (b)) [13]. Note that the two sets of measurements have been performed along different directions, $\langle 110 \rangle$ and $\langle 100 \rangle$, i.e., $\overline{1}\overline{X}$ and $\overline{1}\overline{M}$, respectively. Because of the intrinsic isotropy of the surface, both directions can be used for determining the temperature

dependence of the DE peak. In both TOF spectra the largest off-scale peak is the diffuse elastic scattering from defects, labelled as DE. The inelastic peaks, labelled as RW (Rayleigh wave), or O_1 , O_2 , O_3 , and O_4 are, in that order, much less intense. Recently inelastic neutron scattering (INS) has been used by Merritt et al. [18] to measure the low-energy longitudinal acoustic (LA) and two longitudinal optical (LO) bulk modes of optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$.

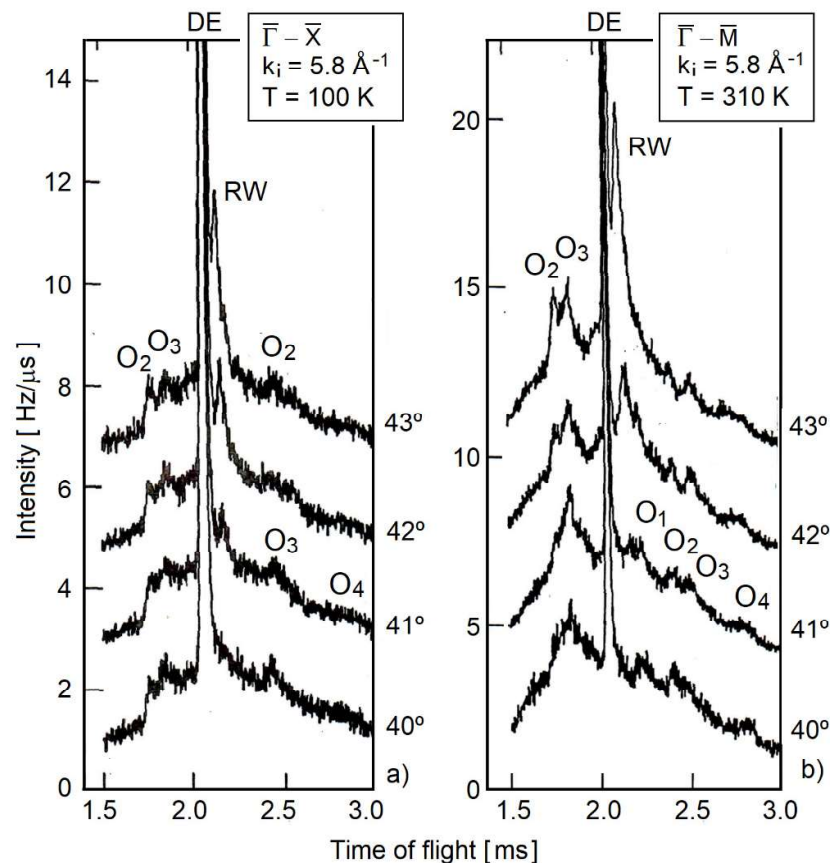


Figure 1. HAS time-of-flight spectra from the Bi-O(001) surface of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, at about the optimal doping condition. The measurements were performed at different incident angles (indicated on the right side of the spectra) with a 90° planar scattering geometry and incident He atom momentum of $5.8 \text{ \AA}^{-1}$: (a) in the $\bar{\Gamma}\bar{X}$ symmetry direction at a surface temperature $T = 100 \text{ K}$, and (b) in the $\bar{\Gamma}\bar{M}$ symmetry direction and $T = 310 \text{ K}$. Besides the diffuse elastic (DE) and Rayleigh wave (RW) peaks, various inelastic peaks, labelled O_1 to O_4 and assigned to phonon creation (right side) and annihilation (left side) are observed (adapted from [13]).

Figure 2 displays the HAS dispersion curves of the surface phonons (open squares), derived from TOF spectra like those of Figure 1 and the INS data (red symbols) along the two main symmetry directions $\bar{\Gamma}\bar{X}$ and $\bar{\Gamma}\bar{M}$. The dispersion curves of the lowest acoustic branch corresponding to the RW and the other low-energy surface phonon branches, O_1 , O_2 , O_3 and O_4 , are clearly delineated. The branches O_2 , O_3 and O_4 are almost dispersionless. At the center zone of $\bar{\Gamma}$, the surface modes O_1 , O_2 , and O_3 appear to be a somewhat softened version of the two bulk LO modes, a possible effect of surface localization. Moreover, the branches O_2 , O_3 , and O_4 look rather similar along the two symmetry directions, also suggesting some sort of isotropy.

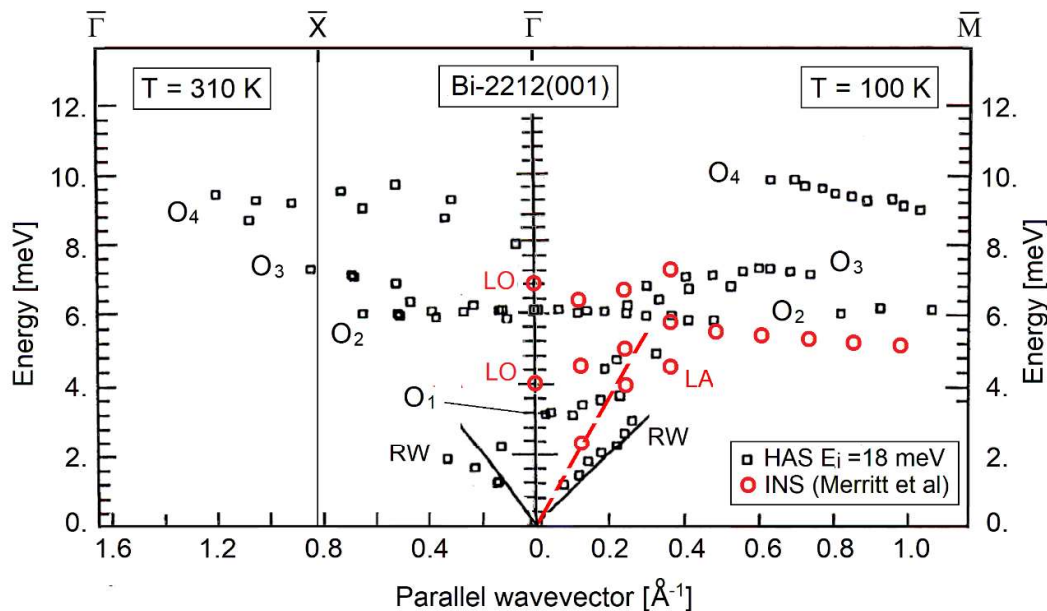


Figure 2. Low-energy phonon dispersion curves at the Bi-O(001) surface of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) with $\delta = 0.16$ at about the optimal doping condition, measured with HAS [13] (open squares) along two different symmetrical directions and surface temperatures (cfr. Figure 1), compared to INS data by Merritt et al. (red open circles) [18]. The apparent isotropy of the O_2 , O_3 and O_4 phonon branches suggests their association to the non-stoichiometric oxygen randomly distributed in the four Bi-O and Sr-O planes within the first half unit cell (corresponding to one published formula [15]).

The present experimental nearly dispersionless curves for the optimally doped material are in contrast to the theoretical large frequencies and strong dispersion of the surface phonons calculated with the shell model for the stoichiometric ($\delta = 0$) compound [19,20]. This suggests that the localized vibrations of random non-stoichiometric weakly interacting oxygen atoms (as neutral acceptors) have a sufficiently high concentration in the four BiO and SrO layers within the first half-unit cell (corresponding to one published formula [15]) and large vibrational amplitudes to exhibit flat and soft dispersion curves. This also indicates that atomic vibrations in the second BiO layer, the seventh layer beneath the surface, are detected. The present interpretation appears to be preferable to the one suggested in previous works [13,20] in which parts of the observed branches have been associated with folding effects from the stripe superstructure, despite the apparent isotropy.

3. Electron–Phonon Interaction from the Temperature Dependence of the Diffuse Elastic Peak

Figure 3 reproduces the TOF spectra of Figure 1 on a reduced ordinate scale (different in panels (a) and (b)), so as to show the decrease in the DE peak intensity with increasing temperature from 100 K (a) to 310 K (b) for the four different incident angles. As seen from the values reported in Table 1, the intensity decrease at 43° is less pronounced than for the other three incident angles. This is attributed to the close proximity to the elastic specular peak at 45° and its intensity tail due to the finite experimental energy resolution. The fair consistency of the decrease rates for the other three incident angles suggests considering λ and its standard deviation derived from the 40° , 41° , and 42° data as more reliable than those derived from averaging over all four incidence angles.

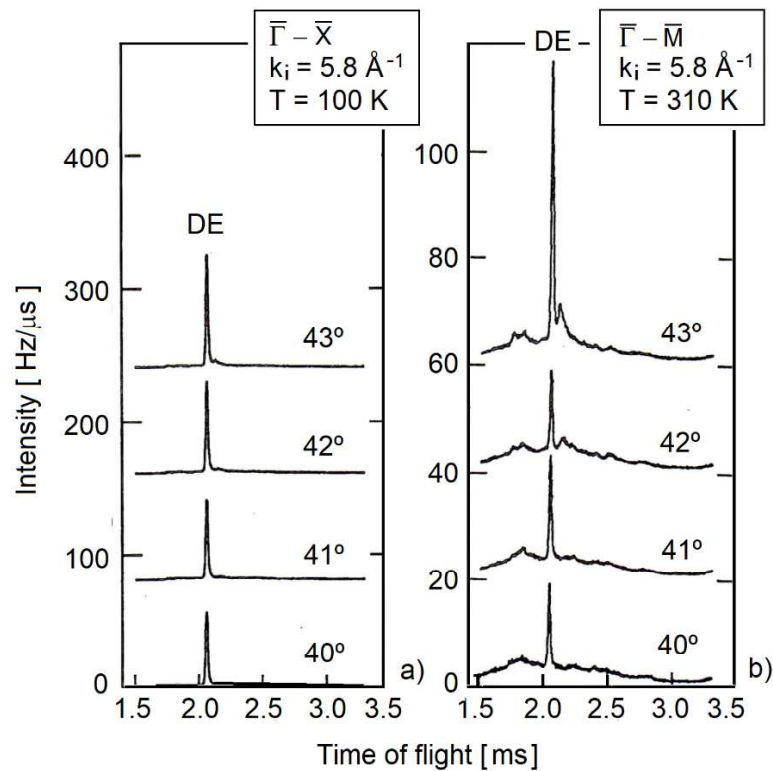


Figure 3. Comparison of the diffuse elastic (DE) peak intensities shown in Figure 1 on a larger ordinate scale measured at two different surface temperatures of 100 K and 310 K for the four incident angles. Note the reduced ordinate scale compared to Figure 1 and the differences in ordinate scale for $T = 100$ K (a) and 310 K (b).

Table 1. HAS diffuse elastic (DE) peak intensities at different incidence angles θ_i and corresponding logarithmic variations with temperature.

θ_i [Degrees]	I_{DE} (100 K) [Hz/ μ s]	I_{DE} (310 K) [Hz/ μ s]	$\Delta \ln[I_{DE}(T)]$
43°	83.3	49.5	0.52
42°	68.8	15.0	1.52
41°	58.3	20.0	1.07
40°	54.5	15.5	1.25

The EP coupling constant (mass-enhancement factor) can be expressed as in Ref. [11], Equation (7), by adapting it to the case of the DE peak:

$$\lambda = \frac{2\pi\phi}{n_s a_c (k_{fz} - k_{iz})^2} \left| \frac{\Delta \ln I_{DE}(T)}{k_B \Delta T} \right|. \quad (1)$$

here, $\phi = 4.85$ eV is the work function of Bi-2212 at 85 K [21]; $n_s = 4$ is the number of layers hosting non-stoichiometric oxygen in the first half unit cell; $a_c = a^2 = 14.55 \text{ \AA}^2$ is the surface unit cell; $k_{iz} = -k_i \sin \theta_i$ and $k_{fz} = k_i \sin \theta_f$ are the incident and final components of the He wavevector normal to the surface; and $I_{DE}(T)$ is the DE peak intensity, as given in Table 1 at the two temperatures.

With these input values and by averaging over $\theta_i = 42^\circ$ to 40° , it is found that $\lambda = 0.55 \pm 0.08$, with the error expressed by the standard deviation. Note that by averaging over all four θ_i values, one would obtain $\lambda = 0.47 \pm 0.16$. Both these values compare well with those reported by Chia et al. (0.46 ± 0.03 from ultrafast optical techniques at the

optimal doping condition and at room temperature) [14], by Lanzara et al. (0.59 ± 0.35 from ARPES at the optimal doping condition, at both $T < 20$ K and > 100 K) [4,5], and by Ino et al. ($\sim 0.5 > \lambda > \sim 0.3$ for $0.1 < \delta < 0.2$ from ARPES at ~ 10 K) [22]. These values of λ are within the same range above and below T_c but are much smaller than the values (> 2) that would be required by EP interaction alone in order to account for a T_c as large as 91 K [23].

4. Conclusions

The method of measuring the EP interaction constant (mass-enhancement factor) at crystalline conducting surfaces by means of specular He atom scattering [8,9], has been extended to non-stoichiometric materials, where the specific contribution of doping to the EP interaction is contained in, and extracted from, the temperature dependence of the diffuse elastic peak. This new concept has been applied to HAS spectroscopy data for the non-stoichiometric high-temperature superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ with $\delta = 0.16$, which is about the optimal doping condition and corresponds to the critical temperature $T_c \sim 91$ K. The value of λ obtained in this way is in good agreement with the values recently estimated with other methods. This confirms that EP interaction, although not solely responsible for high- T_c superconductivity, nevertheless plays a relevant role in this class of layered superconductors. Moreover, EP interaction occurs prior to small polaron formation, and the onset of polaron stripes coexisting with itinerant carriers in doped perovskites [24–26], and the pseudo-Jahn–Teller polaron–polaron attraction has been suggested as a possible pairing mechanism [24,25]. The charge ordering and related stripe superlattices [25,26], as well as the small-polaron spin-moment texture predicted by Koizumi et al. [9] should all be within the high resolution limits of, and therefore accessible to HAS diffraction. Considering the comparatively expedient way of obtaining, with He atom scattering, the total EP interaction constant λ , it is hoped that the present experimental method will be soon applied to quasi-two-dimensional doped superconductors and other low-dimensional non-crystalline nanostructured materials.

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