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Low-temperature phonon damping in vitreous silica explored by UV Brillouin spectroscopy

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ABSTRACT

The recently developed technique of Brillouin ultraviolet scattering is used to measure phonon dispersion and damping in the prototypical strong glass v-SiO_2. Results on the low temperature, T ~ 10 – 100 K, sound attenuation at a frequency around 66 GHz are presented. The comparison between a model which takes into account dynamical processes (thermally activated relaxations and the interaction with the bath of thermal vibrations) and the experimental data indicates the presence of a strong *static* contribution in the region explored by ultraviolet radiation supporting the presence of a transition between *static* and *dynamic* attenuation mechanisms at a frequency around 100 GHz.

1. INTRODUCTION

The study of the low-energy vibrational dynamics in glasses is one of the most intriguing challenges for the understanding of the effects of the topological disorder in solids. Ultrasonic measurements (see [1] and references therein) at low frequency ($\nu < 1$ GHz) and Brillouin scattering [2] at higher frequencies ($\nu > 10$ GHz) give access to the experimental determination of the velocity and damping of collective excitations in glasses. Thus these techniques result the most direct probes available for the study of the low-energy tail ($\nu < 1$ THz) of the vibrational spectrum in amorphous systems.

The determination of the frequency and temperature dependence of the sound attenuation has a role of primary importance for the understanding of the nature of the low-frequency excitations in glasses. This particular frequency region, lying at and below the Boson peak, is practically inaccessible to actual numerical simulations on realistic classical systems [3,4] and for this reason the nature of the corresponding eigenmodes is still uncertain. Different authors [5,6] suggest that both perturbed plane waves, following the scheme proper of the acoustic modes of crystals, and some quasi-localized modes, associated to small values of the participation ratio, could coexist at and below the Boson peak frequency. Moreover, the mean free path of the acoustic waves is strictly related to the transport properties of the system, such as the thermal conductivity. Its temperature

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dependence is one of the main open problems in the physics of structural glasses, in spite of the great amount of work on the subject (see, for example, [7] and [8]).

In ultrasonic measurements (US) as well as in Brillouin light scattering (BLS) the sound attenuation is found to be strongly temperature dependent showing, almost universally, a broad maximum below the glass transition temperature. The position of the maximum increases smoothly and its amplitude increases almost linearly with increasing frequency. Such behaviour is generally understood in terms of a thermally activated relaxation process with the relaxation time following an Arrhenius type activation law [9].

Vitreous silica, considered the prototype of strong glasses and a model for all glasses in general [10] is probably the most widely studied glass in the literature. In particular the topic of sound attenuation has been the subject of a great amount of experimental work accomplished with different techniques (see ref. [11-16] to mention only the most recent ones). In v-SiO_2 at ultrasonic frequencies (MHz) the attenuation, as a function of temperature, shows a well-defined peak, while at hypersonic frequencies (GHz) this temperature dependence, although still characterized by a maximum located at around 150 K, clearly shows a plateau at higher temperatures. The wave vector dependence of the attenuation at room temperature in the region of frequency explored by BLS and Brillouin ultraviolet light scattering (BUVS) has been recently observed [16] to scale with the square of the frequency and thus with the square of the wavevector, being the dispersion curve linear in this region. A q^2 dependence of the attenuation was observed also at much higher frequencies ($\nu \sim$ 1-10 THz) by means of inelastic X-rays scattering (IXS). Measurements of the sound attenuation by IXS showed the absence of any temperature dependence in the explored temperature range [17], thus suggesting a *static* nature for the attenuation in this region.

Recent BUVS room temperature measurements [18] suggest that the transition from the *dynamic* to the *static* attenuation regime should occur at around 100 GHz. The presence of such a crossover should be directly revealed in the temperature dependence of the sound attenuation, which should progressively be reduced going from the frequency region explored by BLS up to the crossover frequency.

In this paper we will present measurements of the sound attenuation in vitreous silica at low temperatures performed with the high-resolution ultra-violet spectrometer called HIRESUV present at the University of L'Aquila [19]. The frequency and damping of longitudinal phonons of v-SiO_2 at the wavevector $q \sim 0.071$ nm^{-1} have been measured in a temperature range from ~10 to 100 K. We will then compare the experimental results for the acoustic attenuation with the prediction of a phenomenological model which takes into account aside the relevant mechanisms of *dynamical* nature in the explored temperature range, namely the thermally activated relaxation processes [20] also the interaction with the thermal vibrations [21]. Our purpose will be to show how the comparison between the expected behaviour and the measured sound attenuation will indicate the presence of a relevant *static* contribution in the ultraviolet frequencies, contribution which is, on the contrary, almost negligible in the region explored by BLS.

2. EXPERIMENT AND RESULTS

The newly developed technique of Brillouin ultraviolet light scattering (BUVS) is used to explore the frequency region between that accessed by BLS and IXS. In this work the spectrometer HIRESUV is used with excitation line of 266 nm and in near backscattering configuration, 173° of scattering angle, in v-SiO_2, corresponding to $q \sim 0.071$ nm^{-1}. The instrument is based on a double *f* 1:20 home-made monochromator of 4 meters focal length specifically designed for Brillouin spectroscopy with both visible (532 nm) and ultraviolet (266 nm) excitations; a detailed description of the instrument can be found in ref. [19]. The experiment is performed on a spectrosil sample purchased by SILO (Firenze). The sample is characterised by an OH^- content of the order of 1500

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ppm, that we determined by means of infrared absorption, consistent with similar values found in the literature for the spectrosil samples [22].

The longitudinal and transverse sound velocities, $v_{L,T}$, are determined from the measured frequency shifts ν_L and ν_T by following formula:

$$v_{L,T} = \frac{2\pi\nu_{L,T}}{q} \tag{1}$$

where q is the exchanged wavevector fixed by the experimental geometry and by the wavelength of the incident radiation. The damping of the acoustic waves is obtained from the spectral width, full width half maximum, Γ, of the Brillouin lines. Assuming that the isotropic contribution to the light scattering signal is dominating the spectrum, the spectra are proportional to the dynamic structure factor, $S(q,\nu)$, convoluted to the resolution function $R(\nu)$, according to the standard theory of Brillouin scattering in glasses [23]. The dynamic structure factor, as derived from a memory function approach to the dynamics of the density fluctuations in the Markov approximation, can be described as a damped harmonic oscillator (DHO) for the inelastic contribution and a delta function for the elastic scattering [24].

In the region of frequencies below 10 GHz the peak of attenuation is found to scale almost linearly with frequency so that the internal friction, $Q^{-1} = \Gamma/\nu$, is the natural quantity to deal with. Moreover it is a dimensionless quantity and it has an immediate physical meaning (for example, $Q^{-1} \approx 1$ defines the Ioffe-Regel regime[‡], where the attenuation is comparable to the frequency).

An example of spectrum, Stokes line, obtained at T ~ 10 K is reported in figure 1 together with the best fitting function and the scaled elastic line used as resolution. Particular attention has been adopted during the fitting procedure to take into account the weak residual presence of the scattering signal due to scattering from the v-SiO_2 windows of the cryostat itself, term growing in importance in lowering the temperature of the sample. This term appears as a small bump on the high frequency side of the Brillouin peak. The extremely high contrast of the spectrometer (up to 10^{10}) may be appreciated in the completely flat background which characterizes the spectrum. The variation of the sound velocity v_L as a function of temperature is reported in figure 2, and appears in good agreement, within experimental error, with previous visible and ultrasonic data. Small differences between the ultrasonic data (literature data from [9]) may be associated to the different thermal history of the silica samples.

In figure 3 the internal friction, Q^{-1}, calculated from the BUVS data, together with the ones of visible measurements taken by ref. [12] and ultraviolet data obtained at the IUVS beam line in Trieste [15] are reported. The difference between the visible and ultraviolet data is evident and supports the idea that in the region of frequencies explored by ultraviolet excitation another attenuation mechanism is summed to the *dynamical* contribution.

The temperature dependence of the internal friction corresponding to five ultrasonic frequencies (11.4 KHz, 180 KHz, 21 MHz, 207 MHz and 748 MHz) is shown in figure 4 together with the BLS measurement. The plot shows that the attenuation peak undergoes a variation of around a factor five in a window of more than *six decades* in frequency, i.e. Q^{-1} is almost constant, up to the region investigated by BLS.

3. DISCUSSION

It is well established that the sound attenuation in the US region (< 1 GHz) in vitreous silica is dominated by thermally activated relaxation processes (TAR), although their microscopic origin is

[‡] Different definitions of this limit can be found in the literature [25], such as $Q^{-1} \approx 1/\pi$ or $Q^{-1} \approx 1/2\pi$.

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still debated. We have analysed the US data using the following model for the TAR contribution (see [26] and references therein):

$$Q_{TAR}^{-1}(\omega,T) = C\Phi\left(\frac{\sqrt{2}T}{\sigma_\Delta}\right)\frac{1}{T}\int_0^\infty\left(\frac{V}{\sigma_V}\right)^{-\zeta}\exp\left(-\frac{V^2}{2\sigma_V^2}\right)\frac{\omega\tau_0\exp(V/T)}{1+\omega^2\tau_0^2\exp(2V/T)}dV \qquad (2)$$

where $\Phi(x)$ is the error function and $\omega = 2\pi\nu$.

Five parameters enter the model of equation (2): the minimum relaxation time, τ_0, two parameters describing the distribution of activation energies, σ_V and ζ, the variance of the distribution of asymmetries in the double well potentials, σ_Δ, and a multiplicative constant C. Assuming a uniform distribution of asymmetries, as usually done in the literature (see [20] and references therein), we fixed the parameter σ_Δ at a value much greater than σ_V, i.e. with $\sigma_\Delta = 10^4$ K. The remaining four parameters can be determined by fitting the temperature dependence of the measured Q^{-1} at a given frequency. A more accurate estimate of the parameters can be obtained by a simultaneous fit of the data at different frequencies.

By the fitting procedure to the five US sets of data presented in figure 4, with σ_Δ fixed at 10^4 K, one obtains the following set of parameters: $\tau_0 = 1.3\cdot10^{-14}$ sec, $\sigma_V = 770$ K, $\zeta = 0.39$ and C variable between 0.17 and 0.19. The result of the fitting procedure on the five sets of US measurements and the extrapolation to the BLS, $\nu = 35$ GHz, data is shown in figure 4. In the plot the multiplicative constant C has been fixed to 0.19 for all the curves. The remarkable good agreement between the model and the ultrasonic measurements can be appreciated, both for the position of the peak of attenuation, which is well reproduced in all the US curves, and also for the intensities, although small discrepancies can be seen. The five US frequencies have been chosen because they are almost linearly displaced in a logarithmic scale and because of the good quality of the data. These sets of data have been obtained in the past by different authors [12,20,27-29] with slightly different ultrasonic techniques and with different samples, so that a certain discrepancy between the model and the data would not be surprising.

The good agreement between the model and the US data demonstrates that a uniform distribution of asymmetries in the double wells can be assumed. With this assumption the model can reproduce the position of the peak in the BLS data but still underestimates the internal friction at this frequency, in particular it does not describe its high-temperature tail. The discrepancy between the TAR model and the BLS data can be associated to the anharmonic contribution, Q_{ANH}^{-1}, arising from the interaction between the acoustic wave and the bath of thermal phonons. It is quite natural that this term results stronger at higher temperatures where the amplitude of the atomic displacements is higher.

A detailed description of the anharmonic contribution, in the Akhiezer formulation, would require a numerical treatment, as discussed in [21]. Unfortunately a numerical approach would suffer from the limited dimensions of the sample that can be modeled on a computer. This difficulty would affect, in particular, the contribution coming from the low frequency tail of the thermal phonon distribution. In the absence of a more general theory describing the anharmonicities, a tentative description of this contribution to sound attenuation is obtained assuming that the effect of the bath of thermal phonons on the acoustic wave can be described by a single relaxation time $\tau_{th}(T)$, which depends on temperature. The anharmonic contribution to the internal friction can then be modeled as [2]:

$$Q_{ANH}^{-1}(\omega,T) = AT\frac{\omega\tau_{th}(T)}{1+\omega^2\tau_{th}^2(T)} \qquad (3)$$

The value of the thermal relaxation time $\tau_{th}(T)$ and its temperature dependence have to be determined from the experimental data. At a fixed frequency Ω, the anharmonic term of eq. (3),

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divided by temperature, is peaked at that temperature T_m for which $\Omega\tau_{th}(T_m) \approx 1$. If this temperature lies in the region explored by the experiment, it is possible to determine $\tau_{th}(T)$ by comparing equation (3) to the experimental data, because the coefficient A can then be fixed by the value of the peak: $Q_{ANH}^{-1}(\Omega, T_m)/T_m$.

The anharmonic contribution, at a given frequency, can be estimated by subtracting the TAR contribution at that frequency from the experimental data. A small *static* contribution shall also be subtracted, as suggested by the recent observation of a crossover between *static* and *dynamic* contributions at a frequency around 100 GHz [18]. The value of such a *static* contribution in the region explored by BLS ($\nu \approx 35$ GHz) can be estimated of the order of the minimum of Q^{-1} which is observed at temperatures around 5 K (with σ_Δ fixed at 10^4 K).

Once the anharmonic contribution, Q_{ANH}^{-1}, has been determined, the thermal relaxation time τ_{th}, in the temperature range of the experimental data, can be estimated by solving equation (3) as suggested in ref. [26]. Fitting the temperature dependence of the thermal relaxation time with an appropriate function, one obtains a model for the anharmonic term of eq (3) and predictions about its frequency and temperature dependence can be done. The temperature dependence of the thermal relaxation time, τ_{th}, is, however, affected by strong fluctuations at low temperatures. For this reason we have chosen a simple linear fit on the logarithmic scale: $\ln\tau_{th}(T) = a + b\ln T$, with a = -20.8 and b = -1.0. With this description of the thermal relaxation time the BLS data are described, as shown by the continue line in figure 5. In the same figure the extrapolations of the TAR contribution of equation (2) and of the anharmonic term (3) at $\nu \approx 66$ GHz are also presented as a dashed line. It appears that the *dynamical* terms, $Q_{TAR}^{-1} + Q_{ANH}^{-1}$, can account only for one half of the sound attenuation measured with the HIRESUV spectrometer.

On the contrary if a *static* contribution $Q_S^{-1} = 0.004$ is added to the *dynamical* terms, then a qualitatively good agreement between UV data and the model can be appreciated, as shown by the dash-dotted line in figure 5. This value for the *static* term, here introduced to describe the UV data, is in qualitative agreement with the result of recent measurements at around 100 GHz [18].

It should however be noted that the OH^- content may have a significant influence on the shape and intensity of the attenuation peak found in the temperature dependence at around 150 K, as pointed out in a recent paper [22]. Nevertheless, the OH^- content does not affect the attenuation for temperatures outside the peak region, below 50 K and above 250 K, where the discrepancy between our UV measurement and the model (dashed line of figure 5) is evident.

4. CONCLUSIONS

We presented new Brillouin ultraviolet light scattering data in the low temperature range in v-SiO_2. We compared the data to a model accounting for both thermally activated relaxations and the interaction with the bath of thermal vibrations. The extension of the model to the ultraviolet region shows that the *dynamical*, or anharmonic, contributions can account for only one half of the observed internal friction, supporting the idea of the existence of a strong *static* contribution in the region explored by ultraviolet spectroscopy.

If the *static* term is included, as in the dash-dotted line of figure 5, the resulting description seems to be in qualitative agreement with the ultraviolet (66 GHz) data that are affected by a relative error of around 20%. This supports the description of the Akhiezer mechanism in terms of equation (3) with a single thermal relaxation time. Further test of equation (3) may be performed,

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for example, by measuring the temperature dependence of the sound attenuation at different scattering angles between the visible and ultraviolet backscattering exchanged wavevectors.

To inspect the nature of this new attenuation term further measurements at low temperature with the HIRESUV spectrometer are in plan.

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FIGURE CAPTIONS

Figure 1: Brillouin spectrum (Stokes side) of vitreous silica at T = (10 ± 3) K in near backscattering configuration obtained with a laser source operating at $\lambda = 266$ nm. The exchanged wavevector is $q = 0.071$ nm^{-1}. The continuous line superimposed to the experimental data (o) is the best-fitted line shape, it is obtained considering only marked circle, as described in the text. The dashed line aligned with the Brillouin peak is the instrumental resolution function. Resolution (F.W.H.M.) = 1.1 GHz, deconvolved line width (F.W.H.M.) = 0.45 GHz.

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Figure 2: Temperature dependence of the sound velocity in vitreous silica, as measured by BUVS, squares (present work), BLS, triangles [12] and US, circles [9].

Figure 3: Temperature dependence of the internal friction ($Q^{-1} = \Gamma / \nu$) in vitreous silica, as measured with IUVS, triangles [15], HiResUv, full circles (present work) and full squares [16], BLS, crosses [26] and dots [20].

Figure 4: Temperature dependence of the internal friction in vitreous silica at different fixed frequencies. Circles: US, 11.4 KHz [20]; squares: US, 180 KHz [27]; products: US, 21 MHz [28]; triangles: US, 207 MHz [12]; stars: US, 748 MHz [29]; crosses [26] and points [20]: BLS, 35 GHz. The continuous lines are the best fitted functions to the five US data sets with the model of equation (2) with the parameter σ_Δ fixed at 10^4 K. Parameters resulting from the fit: $\tau_0 = 1.3 \cdot 10^{-14}$ sec, $\sigma_V = 770$ K, $\zeta = 0.39$ and $C = 0.19$.

Figure 5: Internal friction in vitreous silica at $\nu = 35$ GHz, crosses [26] and points [20] and $\nu = 66$ GHz, circles (present work and [16]). The continuous line is obtained using equations (2) and (3) at 35 GHz, as discussed in the text. The dashed line is the extrapolation of the sum of equations (2) and (3) to the ultraviolet regime, $\nu = 66$ GHz. The dash-dotted line is obtained summing to the dashed curve a *static*, temperature independent, contribution $Q_S^{-1} = 0.004$.

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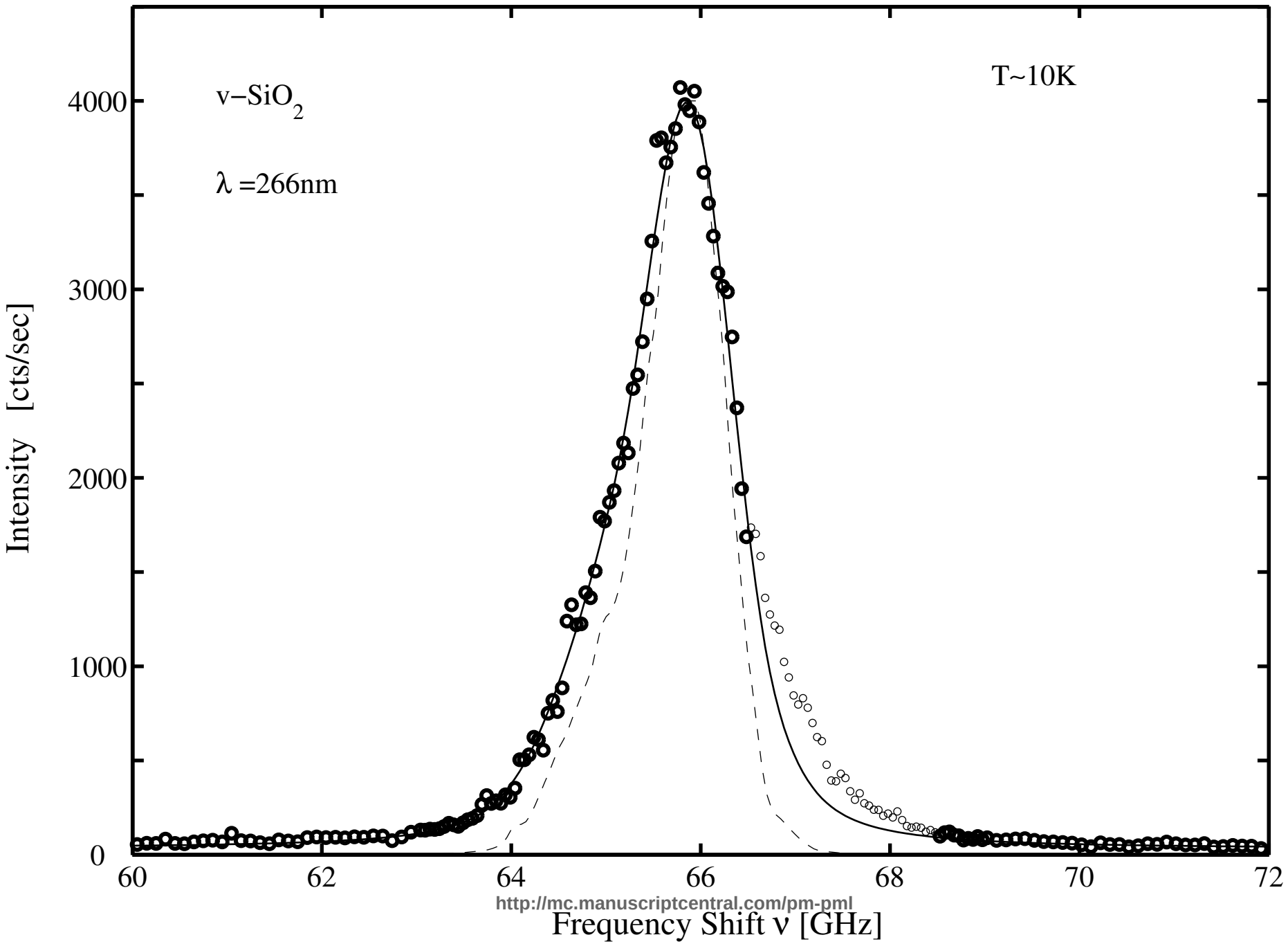
v−SiO2
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T~10K
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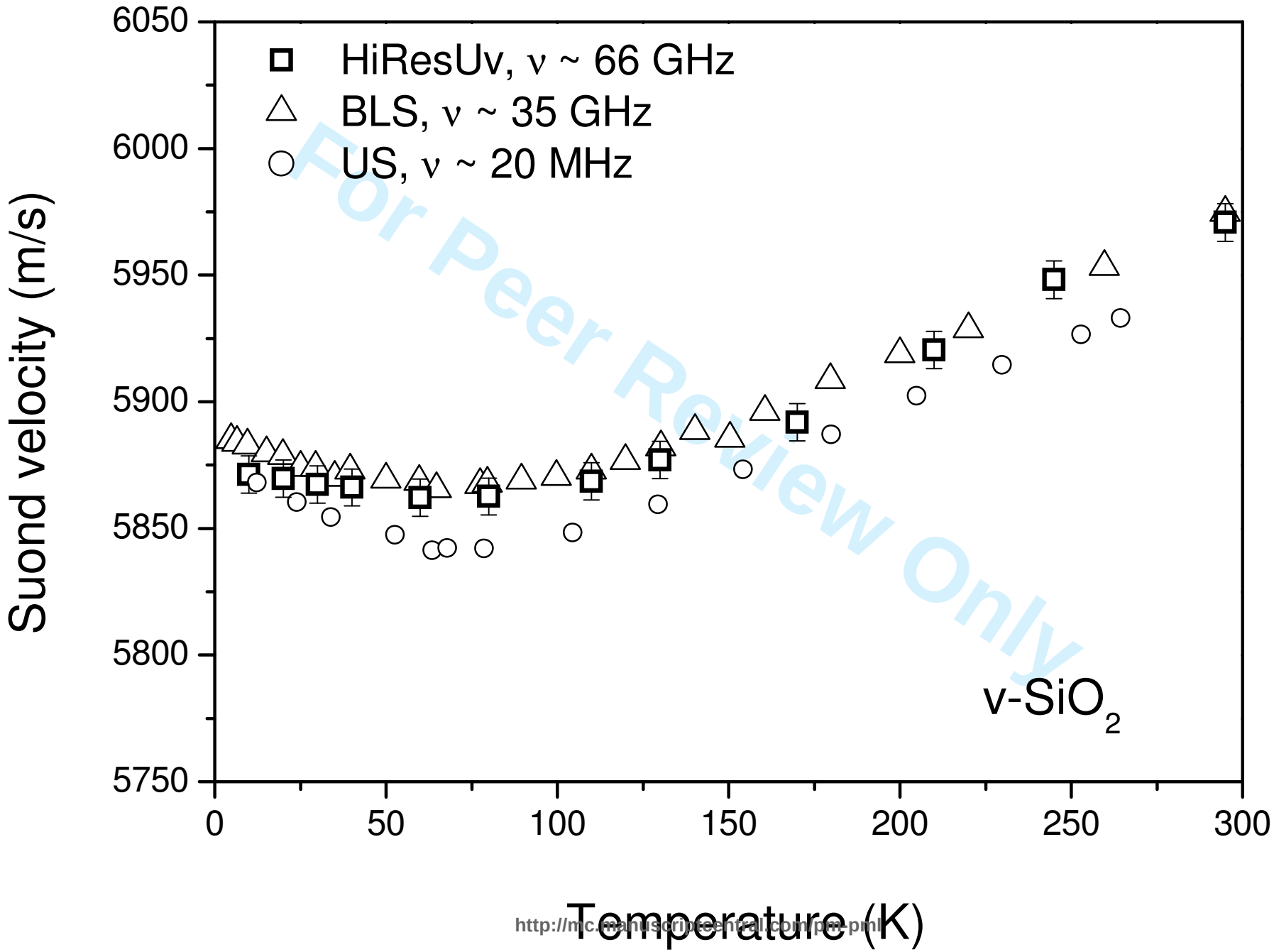
1000
2000
3000
4000
60
62
64
66
68
70
72
Frequency Shift ν [GHz]



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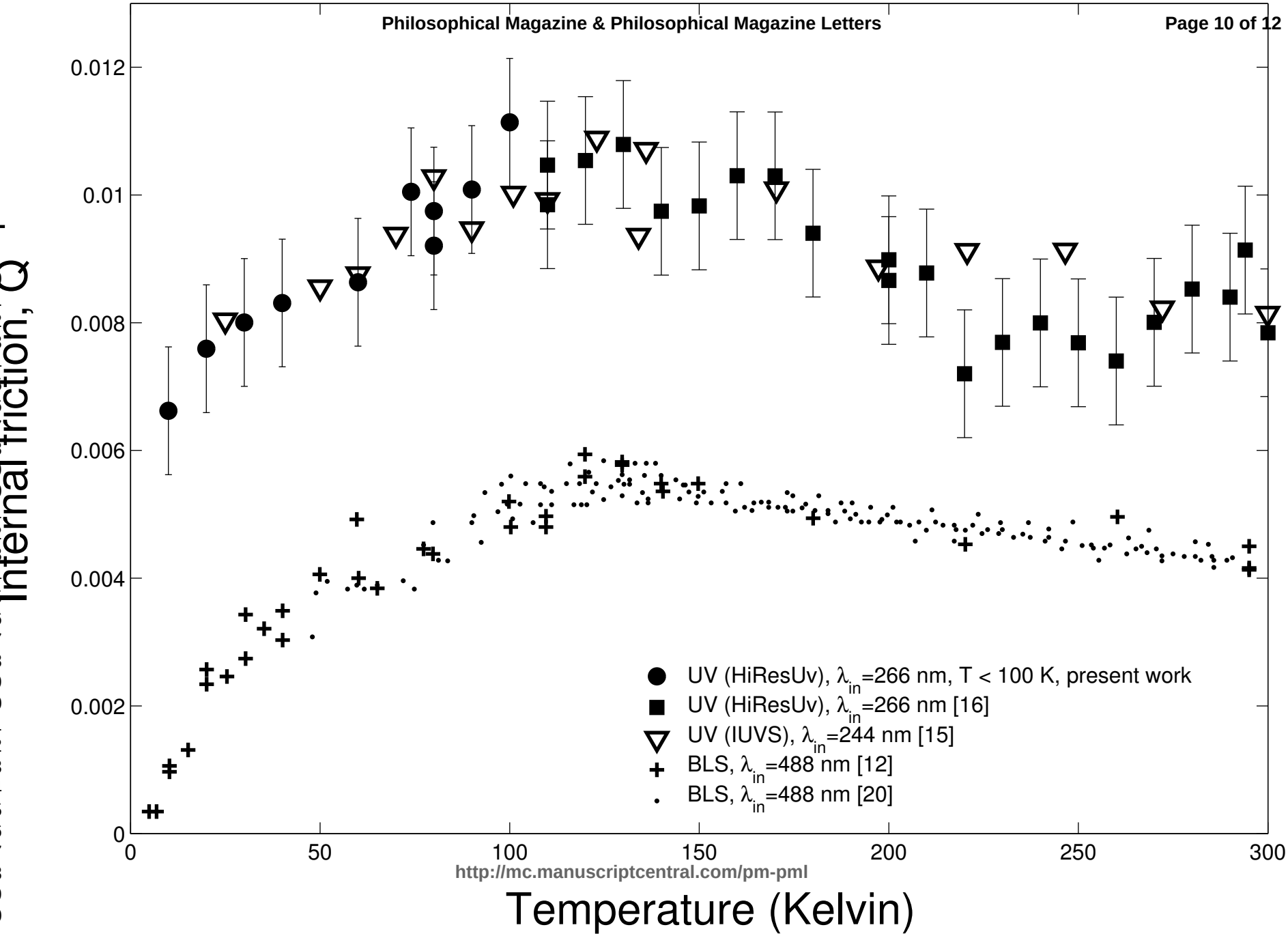
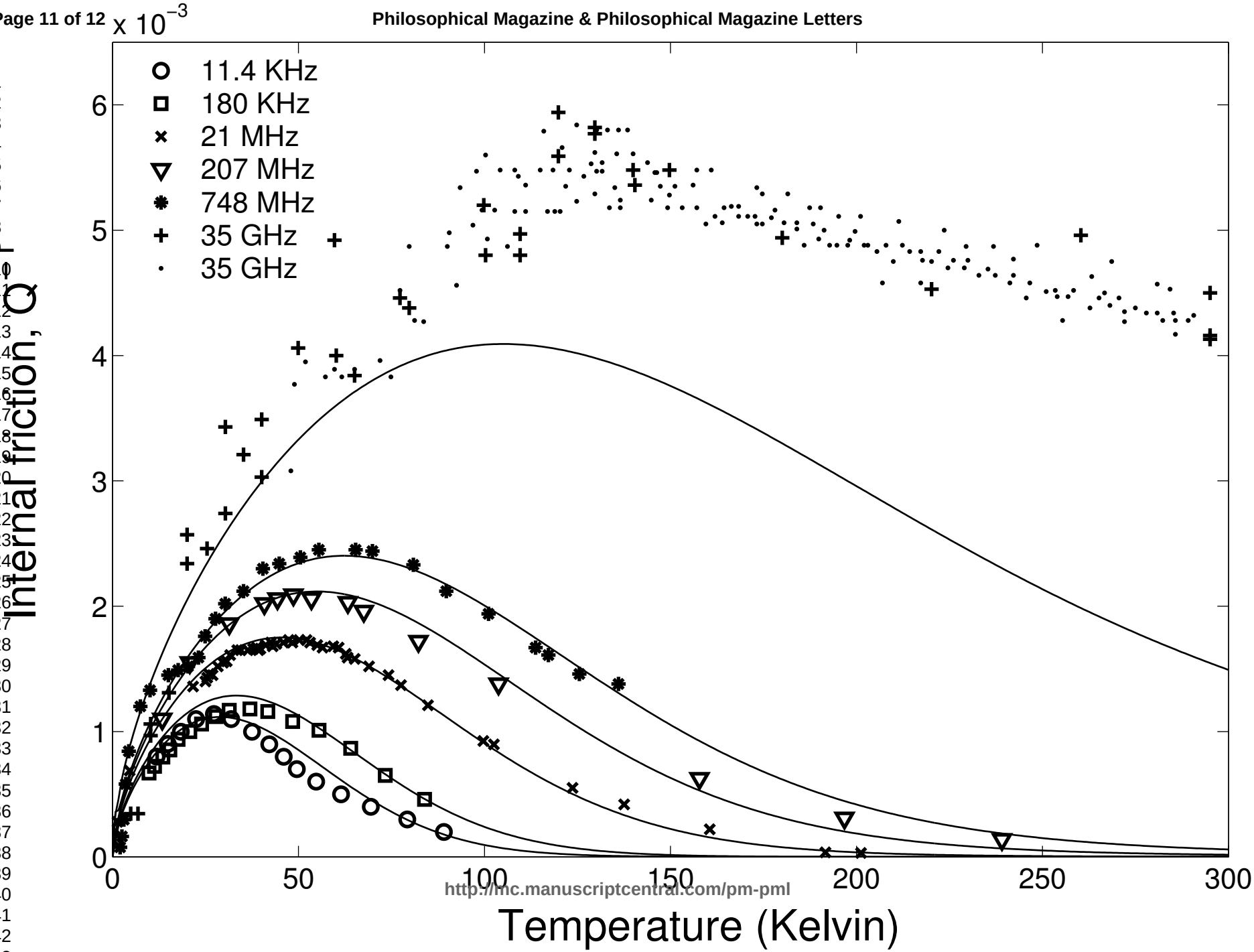

Internal friction, Q^{-1}
0.012
0.01
0.008
0.006
0.004
0.002
0
0
50
100
150
200
250
300
Temperature (Kelvin)
UV (HiResUv), λ_{in}=266 nm, T < 100 K, present work
UV (HiResUv), λ_{in}=266 nm [16]
UV (IUVS), λ_{in}=244 nm [15]
BLS, λ_{in}=488 nm [12]
BLS, λ_{in}=488 nm [20]


x 10^−3
11.4 KHz
180 KHz
21 MHz
207 MHz
748 MHz
35 GHz
35 GHz
Internal friction, Q^−1
Temperature (Kelvin)
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1
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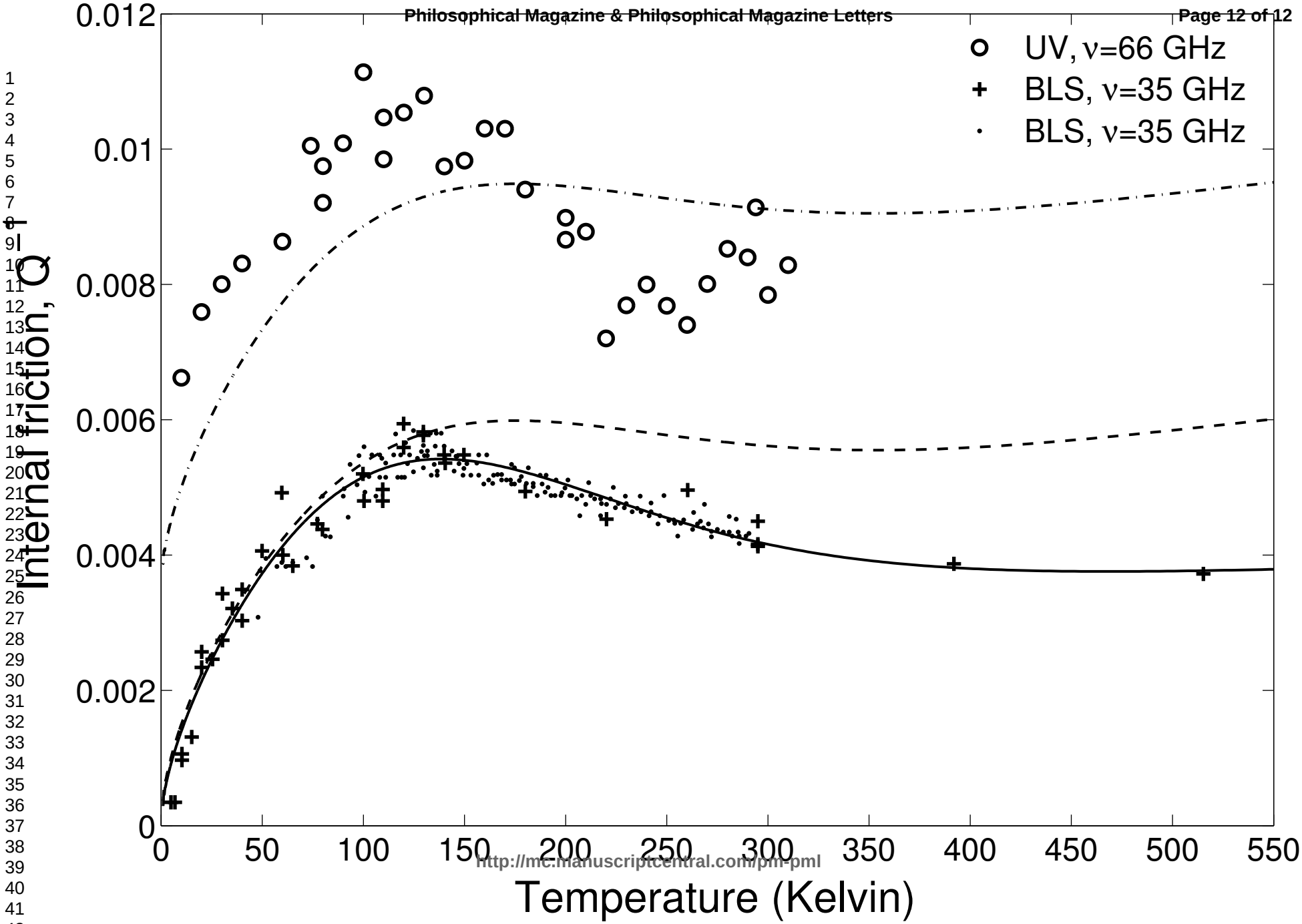
3
4
5
6
0
50
100
150
200
250
300


UV, ν=66 GHz
BLS, ν=35 GHz
BLS, ν=35 GHz
Internal friction, Q⁻¹
Temperature (Kelvin)
0.012
0.01
0.008
0.006
0.004
0.002
0
0
50
100
150
200
250
300
350
400
450
500
550