

# Dislocations in a quantum crystal: Solid helium: A model and an exception

Sebastien Balibar, John Beamish, Andrew Fefferman, Ariel Haziot, Xavier Rojas, Fabien Souris

## ► To cite this version:

Sebastien Balibar, John Beamish, Andrew Fefferman, Ariel Haziot, Xavier Rojas, et al.. Dislocations in a quantum crystal: Solid helium: A model and an exception. Comptes Rendus. Physique, 2016, Condensed matter physics in the 21st century: The legacy of Jacques Friedel, 17 (3-4), pp.264-275. 10.1016/j.crhy.2015.12.015 . hal-01312392

## HAL Id: hal-01312392 https://hal.sorbonne-universite.fr/hal-01312392

Submitted on 6 May 2016

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License



#### Contents lists available at ScienceDirect

## **Comptes Rendus Physique**



CrossMark

www.sciencedirect.com

Condensed matter physics in the 21st century: The legacy of Jacques Friedel

## Dislocations in a quantum crystal Solid helium: A model and an exception

### Dislocations dans un cristal quantique L'hélium solide: un modèle et une exception

Sébastien Balibar<sup>a,\*</sup>, John Beamish<sup>b</sup>, Andrew Fefferman<sup>c</sup>, Ariel Haziot<sup>d</sup>, Xavier Rojas<sup>e</sup>, Fabien Souris<sup>b</sup>

<sup>a</sup> Laboratoire Pierre-Aigrain, Département de physique, École normale supérieure, associé au CNRS et aux Universités Pierre-et-Marie-Curie et Paris-Diderot, 24, rue Lhomond, 75231 Paris cedex 05, France

<sup>b</sup> Department of Physics, University of Alberta, Edmonton, T6G 2E1, Canada

<sup>c</sup> Université Grenoble Alpes, Institut Néel, BP 166, 38042 Grenoble cedex 9, France

<sup>d</sup> Department of Physics, Pennsylvania State University, University Park, PA 16802, USA

<sup>e</sup> Department of Physics, Royal Holloway, University of London, Egham, Surrey TW20 0EX, UK

#### A R T I C L E I N F O

Article history: Available online 17 December 2015

Keywords: Dislocations Elasticity Plasticity Quantum crystals Helium

Mots-clés : Dislocations Élasticité Plasticité Crystaux quantiques Hélium

#### ABSTRACT

Solid helium is paradoxical: it is both a model and an exception. It is a model for crystal properties mainly because of its extreme purity which makes universal phenomena simpler and easier to identify. It is also exceptional because the large quantum fluctuations of atoms around the nodes in their crystal lattice allow these phenomena to occur at very low temperature with a large amplitude. As noticed by Jacques Friedel in 2013, the properties of helium 4 crystals illustrate how the motion of dislocations may reduce their shear elastic modulus, as it does in all hexagonal close packed (hcp) crystals including metals. But this motion takes place without any dissipation in the limit of T = 0 and in the absence of impurities, which is now exceptional and leads to an elastic anomaly at low temperature, which was called "giant plasticity" by Haziot et al. in 2013. More recently, we have discovered that, in helium-4 crystals, helium-3 impurities are not necessarily fixed pinning centers for dislocations. Even at relatively large velocities, dislocations are able to move dressed with impurities somehow as a necklace of atomic pearls through the periodic lattice. This illustrates what is really quantum in these crystals: it is mainly the dynamics of their dislocations and the behavior of impurities.

© 2015 Académie des sciences. Published by Elsevier Masson SAS. This is an open access article under the CC BY-NC-ND licenses (http://creativecommons.org/licenses/by-nc-nd/4.0/).

#### RÉSUMÉ

L'hélium solide est paradoxal : c'est à la fois un cristal modèle et une exception. C'est un modèle pour l'étude des propriétés cristallines à cause de son extrême pureté, qui rend certains phénomènes universels plus simples et plus faciles à identifier. C'est aussi un système exceptionnel, car les fluctuations quantiques de ses atomes autour des nœuds

\* Corresponding author.

E-mail address: sebastien.balibar@lpa.ens.fr (S. Balibar).

http://dx.doi.org/10.1016/j.crhy.2015.12.015

1631-0705/© 2015 Académie des sciences. Published by Elsevier Masson SAS. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

du réseau cristallin permettent à ces phénomènes d'avoir lieu à très basse température, avec une amplitude particulièrement grande. Comme l'avait remarqué Jacques Friedel en 2013, les propriétés des cristaux d'hélium 4 illustrent la manière dont le mouvement des dislocations peut réduire leur module élastique de cisaillement transverse, comme dans tout cristal hexagonal compact (hcp), y compris certains métaux. Mais ce mouvement a lieu sans dissipation lorsque la température tend vers zéro et en l'absence totale d'impuretés, ce qui est exceptionnel et conduit à une anomalie élastique qui a été appelée « plasticité géante » par Haziot et al. en 2013. Plus récemment, nous avons découvert que, dans ces cristaux d'hélium 4, les impuretés d'hélium 3 ne sont pas nécessairement des points d'ancrage fixes pour les dislocations. Même à relativement grande vitesse, les dislocations sont capables de se déplacer habillées d'hélium 3, comme un collier de perles atomiques à travers le réseau périodique. Cela illustre ce qui est vraiment quantique dans ces cristaux : il s'agit principalement de la dynamique de leurs dislocations et du comportement des impuretés.

© 2015 Académie des sciences. Published by Elsevier Masson SAS. This is an open access article under the CC BY-NC-ND licenses (http://creativecommons.org/licenses/by-nc-nd/4.0/).

#### 1. Meeting Jacques Friedel

Sébastien Balibar wishes to dedicate this review article to the memory of Professor Jacques Friedel, with the following personal testimony:

"The first time I mentioned our study of dislocation motion in helium crystals to Jacques Friedel, he said something like "all that has already been measured in hexagonal metals". After a few minutes of surprise, I felt quite happy with that. It meant that our crystals could be of general interest far beyond the little field of quantum solids. So, I insisted and started showing some details of what we were measuring. Together with my visitor John Beamish (Edmonton, Alberta, Canada) and a group of bright students and postdocs who worked on this subject with me at the 'Laboratoire de physique statistique de l'ENS' and are co-authors of this review, we were progressively discovering a series of rather spectacular properties of these dislocations, and I succeeded in capturing Jacques Friedel's attention. From 2011 to the very last moments of his life, we exchanged comments and ideas on the motion of dislocations in crystals. Every meeting was an opportunity to strengthen our growing friendship. These discussions have been extraordinary useful to me. I am extremely grateful to Jacques Friedel for his interest in our work. It is a great honor and a great pleasure for me to summarize our results on the mechanical properties of helium crystals in his memory."

#### 2. Quantum, ultrapure, visible

Why is solid helium-4 considered as a quantum crystal? Above 25 bar and below 1.4 K, <sup>4</sup>He crystallizes in a hexagonal close packed (hcp) structure where the atoms have large quantum fluctuations. This is due to the Heisenberg uncertainty relation between momentum and position. If an atom of mass *m* and diameter *d* is confined by its neighbors in a region of typical size *a*, it acquires a fluctuating momentum  $p = \hbar/(a - d)$  and consequently a quantum kinetic energy  $E_q$  of order  $\hbar^2/[2m(a - d)^2]$ .  $E_q$  is large in the case of <sup>4</sup>He because its mass is low and the hard core d = 2.6 Å is comparable to the lattice spacing a = 3.7 Å. This rough argument leads to  $E_q \approx 15$  K, which is comparable to the depth of the He-He interatomic potential  $V \approx 11$  K [1]. *V* is due to a van der Waals interaction which is rather small because of the low value of the polarizability of He atoms. The large value of quantum fluctuations had already been noticed by F. London in 1936 [2]. It has been recently calculated by E.J. Rugeles et al. [3] who found that the precise magnitude of the kinetic energy associated with quantum fluctuations varies from 10 to 20 K in the liquid, and from 30 to 40 K in the solid, depending on density.

The large quantum fluctuations have a series of consequences. First of all, <sup>4</sup>He remains liquid down to T = 0, with a large molar volume (28 cm<sup>3</sup>/mol). In <sup>4</sup>He gas, the natural concentration of the light isotope <sup>3</sup>He is between 300 and 25 ppb, depending on its precise origin, for example Texas or Qatar wells [4]. <sup>4</sup>He gas can be purified down to the 10<sup>-12</sup> level. <sup>3</sup>He impurities could even be totally removed from <sup>4</sup>He crystals by using an adapted zone melting method [5]. Since all chemical impurities can be filtered out, <sup>4</sup>He crystals are the purest crystals one can study. But, as we shall see below, the elastic properties of <sup>4</sup>He crystals are extremely sensitive to tiny traces of <sup>3</sup>He impurities.

<sup>3</sup>He having a smaller mass, its molar volume in the liquid state is even larger: 37 cm<sup>3</sup>/mol. <sup>4</sup>He crystallizes only above 25 bar (<sup>3</sup>He above 30 to 35 bar, but the crystals under consideration in the present review are <sup>4</sup>He crystals). There is no triple point in the phase diagram where the liquid, the gas and the solid phase would meet as in usual materials. Helium crystals are always grown from their liquid phase. The solid-vapor interface does not exist. Furthermore, around the nodes of their crystalline structure, the atoms fluctuate more than in ordinary classical crystals. For example, the "Lindemann criterion" says that, in most classical crystals on their melting curve, the amplitude of the thermal fluctuations is typically 10% of the interatomic distance [6]. In <sup>4</sup>He crystals, Burns and Isaacs [7] measured it and found a root mean square displacement of 26%. In other words, the He atoms are weakly localized in their network.



**Fig. 1.** The growth shape of helium crystals shows evidence for their hexagonal structure. This picture was taken in 1994 at 0.1 K in their optical refrigerator by S. Balibar, E. Rolley and C. Guthman. From the angle between the basal facet and the 6 prismatic ones, the orientation of the six-fold symmetry axis *c* can be determined. This crystal is 1 cm wide. The colors are obtained by illuminating it with white light dispersed through a prism [8].



**Fig. 2.** The experimental cell that was used to measure the mechanical properties of <sup>4</sup>He crystals at ENS in 2013–2014. Single crystals are grown from the bottom up inside the 0.7-mm slit between two vertical transducers in the center of the cell. The crystal orientation was obtained from photographs of the growth shape in the bottom part.

Given these peculiarities, some authors considered that quantum fluctuations could destroy the surface order leading to the existence of facets on the crystal shape. This was proved wrong [8]. In reality the existence of facets is a consequence of the existence of a periodic potential due to the crystalline structure, which localizes the crystal surfaces near crystal planes at low enough temperature. This potential is reduced by quantum fluctuations but not to zero. The existence of facets has proved essential in all our studies because, thanks to the windows of our refrigerator, we could determine the crystal orientation from photographs of their growth shape such as shown in Fig. 1 [9].

Some other authors proposed that <sup>4</sup>He crystals could be superfluid, not only liquid <sup>4</sup>He. A coexistence of crystalline order in real space and superfluid order in momentum space is called "supersolidity" and is definitely paradoxical but not impossible. An experiment by Kim and Chan [10,11] had raised a lot of interest for this question in 2004. They had observed an anomaly in a "torsional oscillator" (TO) filled with solid <sup>4</sup>He, which they interpreted as a consequence of supersolidity appearing below 0.1 K. However, the resonance period of a TO is proportional to the square root of a momentum of inertia *I* divided by an elastic constant *K*. Supersolidity would indeed imply an anomalous reduction in *I*, but an increase in *K* could have the same effect. Day and Beamish [12] showed in 2007 that <sup>4</sup>He crystals had a larger shear modulus below 0.1 K than above. After several years of studies, it was shown by several groups [13–15] that, in most experiments the rotation anomaly observed in TOs was due to a change in elastic shear modulus, not to any superfluid flow reducing the momentum of inertia. The discovery of supersolidity would have been a major event, but our careful study of the mechanical properties of <sup>4</sup>He crystals led us to the discovery of a "giant plasticity" [16], which is not less spectacular nor less interesting. This plasticity is the subject of the present review.

#### 3. A giant plasticity

To measure the shear modulus of various <sup>4</sup>He crystals, we used successive experimental cells. The last one is shown in Fig. 2. It is made of a roughly hexagonal hole in a copper plate that is attached to a dilution refrigerator, whose temperature can reach 15 mK even when its large windows are open for optical measurements. The cell is closed by two sapphire windows so that an ordinary camera can record growth shapes of the crystals under study [9,16]. It contains two piezoelectric transducers made from PZT material with a transverse polarization. Oriented single crystals of <sup>4</sup>He are grown along the vertical slit (0.7 mm thickness in this case) between the two transducers. One transducer produces a vertical displacement (0.95 Å/V), while the other measures the stress that is transmitted by the <sup>4</sup>He crystal. The sensitivity and the stability of



**Fig. 3.** In a temperature domain around 0.2 K, this ultrapure crystal shows a "giant plasticity": its shear modulus is highly reduced with respect to its intrinsic value (127 bar), which is due to the elasticity of the lattice only. This plasticity is due to the large mobility of dislocations in this temperature domain. At lower temperature, traces of <sup>3</sup>He impurities start binding to the dislocations and preventing their displacement. At higher temperature the motion is damped by collisions with thermal phonons.

this setup are such that one could apply very small strains in the range from  $10^{-10}$  to  $10^{-6}$  and measure stresses down to  $10^{-9}$  bar. The stress to strain ratio gives an absolute measurement of the shear modulus in one particular direction thanks to a careful calibration. Strains are applied at a frequency between 1 Hz and 20 kHz so that, thanks to a lock-in amplifier, both the amplitude of the elastic shear modulus and the dissipation could be measured.

Fig. 3 shows one particular recording of the shear modulus of a crystal that was oriented with its six-fold symmetry axis (the *c* axis of the hcp structure) nearly vertical. In this particular crystal named X15 by Haziot et al. [16], the concentration of <sup>3</sup>He impurities had been reduced to  $4 \times 10^{-10}$  only. In a temperature region around 0.2 K, its shear modulus is 43% smaller than the intrinsic modulus (127 bar) that is known from sound velocity measurements at higher frequency and higher temperature. By intrinsic we mean what is only due to the elasticity of the crystal lattice. But, as explained in all necessary details by Jacques Friedel in his famous book [17], real crystals contain linear defects called "dislocations" whose motion may produce an additional strain under the action of an applied stress. It means a reduction in the shear modulus. We have demonstrated that, in the case of <sup>4</sup>He, highly mobile dislocations strongly soften the crystals but only in a temperature domain between two other regions where two different mechanisms prevent the dislocation lines from moving. It depends on their concentration but typically below 0.1 K, <sup>3</sup>He impurities bind to dislocations and either pin or slow down their motion. As for the region above 0.3 K, it is the collisions with thermal phonons, which slow down this motion.

Before considering this elastic anomaly in more details, let us make a quick comparison with classical crystals. In classical crystals, dislocations move only at high enough temperature and under sufficiently large stress. This is because dislocations are defect lines moving in a periodic lattice, something which may only happen by thermal activation above energy barriers called "Peierl's barriers" [17]. Applying a stress reduces these barriers. High thermal fluctuations allow point defects (named "kinks" or "jogs") on the dislocation lines to pass above the barriers and produce a displacement of the line. In classical crystals, the dislocation motion induces a small softening that is highly dependent on temperature and stress amplitude, while in <sup>4</sup>He it is large and independent of temperature if impurities are removed. We shall discuss below the relevance of the term "plasticity" which some specialists in Material's science consider misleading, but which J. Friedel accepted [18].

Fig. 3 suggests that, at low temperature, the pinning of dislocations by impurities is not yet completed at the lowest temperature we could reach. If due to <sup>3</sup>He impurities, using natural helium with a larger <sup>3</sup>He concentration  $X_3$  should displace the pinning to higher temperature and allow to reach the complete pinning at reachable temperatures. This is indeed what we observed with various crystals of natural purity containing  $X_3 = 2.5 \times 10^{-8}$  <sup>3</sup>He, as shown on Fig. 4. Contrary to the ultrapure crystal X15 of Fig. 3, all the crystals with natural purity (X2, X3, X5, X6, X20, X21 and a polycrystal) reached their intrinsic shear modulus below 60 mK. This intrinsic value is indicated with tics on the vertical axis. It strongly depends on orientation. Note the particular crystal X3, whose *c* axis was tilted by 45 degrees from the vertical: it shows no temperature dependence at all, which allowed us to calibrate our transducers.

The elasticity tensor of hexagonal close packed crystals contains only 5 elastic coefficients. Among them, the coefficient  $c_{44}$  determines the stress that is necessary to make basal planes glide against each other (see cases *b* and *b'* on Fig. 5). The coefficient  $c_{66}$  determines the stress that is necessary to deform the hexagonal symmetry in these basal planes (see case *a* on Fig. 5). What happens in the particular case of X3 is that the response is independent of both  $c_{44}$  and  $c_{66}$ . Our measurements showed that the elastic anomaly is a softening from the intrinsic value when dislocations are pinned (at



**Fig. 4.** The magnitude of the change in shear modulus is highly sensitive to the orientations of crystals with respect to the vertical shear that is used to measure this shear modulus. All crystals are made out of natural <sup>4</sup>He gas that contains  $2.5 \times 10^{-8}$  <sup>3</sup>He impurities, except X15 ( $4 \times 10^{-10}$ ).



**Fig. 5.** Usual notations for stresses and corresponding strains in hexagonal crystals. The case *a* (top line), corresponds to a deformation of the hexagons in the basal planes, where the applied stress  $\sigma$  is proportional to the elastic coefficient  $c_{66}$ . The two cases *b* and *b'* below correspond to the gliding of basal planes with respect to each other, where the applied stress  $\sigma$  is proportional to the elastic coefficient  $c_{44}$ . We are grateful to the anonymous referee who allowed us to reproduce this figure.

low *T*), and that this softening is highly anisotropic. It suggested that it is either due to a reduction in  $c_{44}$  or in  $c_{66}$ , and we have shown that it is  $c_{44}$ , not  $c_{66}$ . Indeed, from the crystal orientation it is possible to express the measured shear modulus in terms of all the elastic coefficients. Then we could either extract the *T*-dependent anomalous value of  $c_{44}$  by assuming that  $c_{66}$  and the three others are constant, or do the opposite, that is assume that  $c_{44}$  is constant and extract a *T*-dependent value of  $c_{66}$ . Fig. 6 shows that the assumption of a constant  $c_{66}$  is correct, since it gives similar values for the reduction of  $c_{44}$  (-60%), while the opposite assumption with a constant  $c_{44}$  would lead to results that are absurd (changes in  $c_{66}$  by more than 1000% for X21 and by 300% for X6). Now, what is the origin of this reduction? It is due to the fact that dislocations



**Fig. 6.** These four crystals have different orientations, but they show the same variation of their elastic coefficient  $c_{44}$ . It has been understood by Haziot et al. as a consequence of the gliding of dislocations that occurs only parallel to the basal planes of the hcp structure (see text).



**Fig. 7.** The relative amplitude of the shear modulus  $c_{44}$  for 4 different crystals at 20 mK as a function of the stress projected on the basal plane ("resolved"). At a threshold stress of a few µbar, dislocations break away from <sup>3</sup>He impurities. This threshold is larger when increasing the stress than when decreasing it. Being free of impurities, the crystal X4 shows a reduction of  $c_{44}$  by 80%, independent of stress.

have preferential gliding directions. A reduction in  $c_{44}$  means that dislocations glide parallel to the basal planes. Thanks to discussions with J. Friedel and O. Hardouin-Duparc, we found a reference by B. Legrand [19] who explains that this is due to the splitting of edge dislocations into two "partial" dislocations with a "stacking fault" in between, if the stacking fault energy is small enough. Real dislocations are often atomic ribbons, not really 1D-lines, in which case they glide parallel to the ribbon plane. In many hexagonal metals (Be, Mg, Co, Zn), gliding is also parallel to basal planes, but in some others (Zr, Ti) it is along prismatic planes. We see that <sup>4</sup>He crystals show phenomena which exist in classical materials but perhaps not as clearly. Indeed, not only the amplitude of the softening is very large in <sup>4</sup>He (we have observed up to 90% reduction in  $c_{44}$  [20]) but, in classical crystals at high temperature, the gliding of dislocations is usually mixed with many other phenomena.

It appeared also possible to remove all impurities from <sup>4</sup>He crystals by a method that is reminiscent of the classical "zone melting". It is based on the fact that impurities are usually more soluble in the liquid than in the solid, where the strain field around each impurity costs elastic energy. In the case of <sup>4</sup>He, the difference in potential energy between the liquid and the solid has been calculated and found to be -1.359 K per <sup>3</sup>He atom [5,22]. As a consequence, when cooling down a crystal in the presence of its liquid phase (remember that there is no triple point in the phase diagram of <sup>4</sup>He), all the <sup>3</sup>He impurities are trapped in the liquid phase. It helps shaking gently the dislocations by applying an oscillating stress so that <sup>3</sup>He impurities do not bind to dislocations and are free to move towards the liquid, as we shall see later. Fig. 7 shows that



Fig. 8. The stress-strain diagram of crystal X4 show a linear behavior with a reduced slope (same data as in Fig. 7). On the contrary, the plasticity of classical crystals is a non-linear phenomenon [21].

at low temperature when impurities are bound to dislocations (crystals X2, X5 and X6), applying an oscillating strain larger than a few microbars detaches the impurities so that the shear modulus is reduced. For crystal X4 where all impurities have been detached before cooling, the shear modulus is reduced by 80% from its intrinsic value and it stays at this low value when cycling the applied stress. Fig. 8 shows that this impurity free crystal has a reduced elasticity that is independent of stress, contrary to classical crystals that would show their intrinsic elasticity at low stress and a reduced one at high stress, which is the classical non-linear plastic behavior. In the case of <sup>4</sup>He crystals the elasticity is reduced by the high mobility of dislocations. That is why we called it a "giant plasticity". But this plasticity is linear, which may be confusing for Materials scientists who are more familiar with classical plasticity, which is also due to the motion of dislocations but is a non-linear phenomenon.

#### 4. The dissipation associated with dislocation motion

In order to understand the dislocation motion, we have measured the dissipation from the phase shift of the crystal response to a driving strain. There are two separate domains in temperature. At high temperature, i.e. above 0.2 K, Granato and Lucke [23] had predicted in 1956 that dislocations should interact with thermal phonons. These interactions would transfer momentum from the dislocations to the phonon gas where the corresponding energy should thermalize. This theory was later improved by Ninomiya [24]. If true, this mechanism should lead to a maximum change  $\delta c_{44}$  in the shear modulus  $c_{44}^0$  that is given by

$$\frac{\delta c_{44}}{c_{44}^0} = \frac{\alpha \Lambda L^2}{1 + \alpha \Lambda L^2} \tag{1}$$

and to a dissipation 1/Q

$$\frac{1}{Q} = \frac{\alpha \Lambda L^2}{1 + \alpha \Lambda L^2} B L^2 \omega T^3$$
<sup>(2)</sup>

where  $\Lambda$  is the density of dislocation lines per unit volume, *L* is a typical length between nodes in the dislocation network,  $\alpha = 0.019$  and B = 905 s m<sup>-2</sup> K<sup>-3</sup> is a thermal phonon damping parameter calculated [4] by using Ninomiya's work [24].

Haziot et al. [26] measured the dissipation 1/Q. Fig. 9 shows a remarkable agreement with theory. The initial slope is perfectly consistent with the predicted  $\omega T^3$  behavior. With a more precise calculation taking care of the existence of a distribution in lengths *L*, Fefferman et al. [25] could fit the whole curve, not only the initial slope. But the rough approximation with a single length *L* already leads an important conclusion. Indeed, fitting our results with eqs. (1) and (2), allows to determine two important quantities, the line density  $\Lambda$  and their mean length *L*. If dislocations formed a regular cubic lattice, these two quantities would be related by the simple relation  $\Lambda L^2 = 3$ . For any three dimensional lattice of dislocations, the density  $\Lambda$  has to be of order  $1/L^2$ . We have found that it is not the case. In their 2013 experiment, Haziot et al. [26] found densities  $\Lambda$  between  $10^4$  and  $10^6$  per cm<sup>2</sup>, which is rather small, and lengths *L* between 100 and 230 µm, which is very large. The latter values are macroscopic and, most interestingly, much larger than for a 3-dimensional network of dislocations. The product  $\Lambda L^2$  ranges from 17 to 57. In an attempt to make even better quality crystals, Souris et al. [20]



**Fig. 9.** Above 0.3 K, the dissipation 1/Q associated with the dislocation option is proportional to  $\omega T^3$  as expected if this dissipation is due to collisions with thermal phonons [23]. The departure from linearity has been understood by Fefferman et al. as a consequence of the distribution of dislocation lengths [25].

found  $\Lambda L^2$  values up to 471! It means that the dislocations do not form a 3D-network. They avoid crossing by forming 2D arrays of parallel lines called "sub-boundaries", and they glide together parallel to the basal planes in a cooperative way.

As can be found once more in his book, Jacques Friedel explains that such a formation of sub-boundaries implies a much larger softening (Souris et al. [20] found up to 90%) than in the case of a 3D network where it is of order 10%. This type of collective motion had been predicted in 1940 by W.L. Bragg [33] and observed in 1952 by Parker and Washburn [34]. Jacques Friedel showed in 1955 that, in aluminum crystals, dislocations are also arranged in sub-boundaries and could move enough near the melting temperature that the shear modulus is highly reduced [27]. He remarks that when approaching the melting point at 933 K, the dissipation associated with the dislocation becomes very large and the dislocation motion mixes with grain boundary migration.

Coming back to our <sup>4</sup>He crystals, our results could be thought of in analogy with a pile of paper sheets that is stiff in all directions except parallel to the sheets were the pile is very soft. But it is not exactly the whole atomic planes which move, it is part of them near each dislocation. Furthermore, there is a non-negligible friction between paper sheets or in classical crystals so that the stress/strain relation is non-linear, while, as shown in Figs. 7–9, the dissipation associated with the shear motion in <sup>4</sup>He tends to zero at low temperature in the absence of impurities. This is an evidence for quantum behavior. One possibility is that quantum fluctuations make the kink energy vanish so that dislocation lines are totally free to move in a periodic lattice that has no influence on them. Another possibility is that kinks have a non-zero energy but dislocation lines contain some kinks for geometric reasons, but these "geometric kinks" would have to move by quantum tunneling through very small Peierl's barriers, a situation that would be hard to distinguish experimentally from the first one.

#### 5. The binding of impurities to moving dislocations

In the above section we mainly focused on the dissipation above 0.3 K where it has been shown to be a consequence of an interaction with thermal phonons. Below 0.2 K, the dissipation mechanism is different. When the <sup>3</sup>He concentration  $X_3$  is not negligible, these impurities progressively bind to dislocations as *T* decreases. A definite dissipation is associated with this binding, as shown by Fig. 10. When impurities start binding, the dislocation motion decreases and the dissipation increases. It reaches a peak at some temperature  $T_p$  before vanishing at low *T* when the dislocations are fully anchored. We studied the frequency dependence of  $T_p$ .

Knowing the typical length and density of dislocations in the crystals under study, Haziot et al. [35] could determine their typical displacement and the speed of their middle point for a given strain. The result is shown on the semi-log plot of Fig. 11 where two different regimes appear. At high speed, the peak temperature is independent of speed. This is the expected regime where impurities act as pinning points: they cannot move fast enough and they anchor dislocations. But at low speed, more precisely below 45  $\mu$ m/s, the constant slope demonstrates a thermally activated regime of friction where the speed decays exponentially as the temperature is reduced and more impurities bind. It means that the dislocations move dressed with impurities attached to them. Assuming that the friction is proportional to the density of bound <sup>3</sup>He, the slope in this semi-log plot allows to determine the binding energy  $E_3$  of <sup>3</sup>He impurities to the dislocation lines. We found  $E_3 = 0.67$  K for this particular crystal. In a more precise study [4], we found a distribution of binding energies around



**Fig. 10.** The temperature variation of the shear modulus (a) and the dissipation associated with dislocation motion (b). Recordings at different frequencies of the driving strain show that the transition from stiff (at low T) to soft (at higher T) depends on frequency in the low-frequency range only.



**Fig. 11.** Knowing the density of dislocation lines and their mean length between the nodes in their network, Haziot et al. [21] could calculate the mean dislocation speed and study its dependence on the temperature  $T_p$  of the peak dissipation in the middle of the transition. Two regimes appear: below 45 µm/s, <sup>3</sup>He impurities move attached to the dislocations while, above this critical speed, dislocations are really pinned by <sup>3</sup>He impurities (see text).



**Fig. 12.** Semilog plot of the relaxation time  $\tau$  of dislocations versus the inverse temperature 1/T for crystals with different orientations and impurity concentrations. Slight variations in slope show that there is a narrow distribution in the binding energy  $E_3$  of <sup>3</sup>He impurities to dislocations (see text).



Fig. 13. Three recordings of the shear modulus measured by Fefferman et al. [25] while decreasing the driving strain near 25 mK. By analyzing the shape of the transition toward the intrinsic shear modulus at low strain, Fefferman determined the width of the distribution of lengths between the nodes of the dislocation network.

0.7 K with a typical width of 0.1 K. This can be seen on Fig. 12 where the relaxation time  $\tau = (1/\omega)\sqrt{1 + \alpha \Lambda L^2}$  show slight variations in slope for different crystals. Our assumption of a dissipation proportional to the concentration  $X_3$  of <sup>3</sup>He impurities was verified by Souris et al. [4] by comparing three types of crystals with  $X_3$  respectively equal to  $2.5 \times 10^{-8}$ ,  $3.8 \times 10^{-7}$  and  $2.32 \times 10^{-6}$ .

A distribution of binding energies was expected because we know that dislocations are rarely purely edge nor purely screw type. Depending on their orientation in the lattice, they have a mixed character so that the binding energy varies slightly from a value corresponding to a pure screw type to a pure edge type where it should be slightly larger. As we shall see now, we have also found some evidence for the existence of this energy distribution from other fits with theory. Indeed, we found evidence for a distribution in the "network length" between nodes in the dislocation network by studying the strain dependence of the shear modulus at low temperature. If one applies a large oscillating strain amplitude  $(10^{-6})$  at 0.5 K and cool down, the large oscillations of the dislocation lines prevent the impurities from binding. When reducing then the strain amplitude, <sup>3</sup>He impurities start binding and Fefferman et al. [25] showed that the shear modulus starts increasing (see Fig. 12). If there were a single pinning length, there should be a precise value of the applied strain at which all dislocations would get pinned and the shear modulus should jump to the intrinsic value. The smooth transition one sees on Fig. 13 is an evidence that it is not the case. In fact the shape of the transition from soft at high strain to stiff at small strain allowed Fefferman to determine the length distribution. This is because short dislocations move less than long ones, <sup>3</sup>He impurities bind to the short ones before long ones as the driving strain is reduced. The network length distribution was found to be rather broad, extending typically from 30 to 300 µm in the case of this particular crystal.

When trying then to fit data sets similar to those shown in Fig. 10, that is recordings of the stiffness and of the dissipation as a function of temperature, it appeared necessary to account not only for a distribution of network lengths but also of binding energies of <sup>3</sup>He. In doing this, Fefferman et al. [25] found that the width of this energy distribution had to be of order 0.1 K around 0.7 K, in very good agreement with the strain dependence in Fig. 13.

#### 6. Comments and open questions

Our discovery of a regime where <sup>3</sup>He impurities move attached to dislocation lines deserves some comments. In a quantum crystal such as <sup>4</sup>He, it has been shown by NMR studies that, at low enough temperature and concentration  $X_3$ , <sup>3</sup>He impurities move by quantum exchange with their neighboring <sup>4</sup>He atoms [28–30]. This tunneling from site to site being coherent, <sup>3</sup>He atoms behave as quantum quasi-particles moving freely through the lattice at velocities in the range form 0.6 to 12 mm/s. This very large quantum mobility explains why, in the presence of a liquid phase in the experimental cell, these impurities get trapped in the liquid, they do not stay in an out of equilibrium state inside the solid. We understand that <sup>3</sup>He atoms attached to dislocation lines cannot move as fast (45 µm/s) as free <sup>3</sup>He atoms because, near dislocations, the lattice is strained so that quantum exchange with <sup>4</sup>He is at least perturbed. As for the mechanism that explains the observed dissipation, we proposed with the help of H.J. Maris [4] that it is an emission of transverse waves along the dislocation lines.

Our series of measurements of stiffness and dissipation of  ${}^{4}$ He crystals as a function of orientation, temperature, driving strain, and  ${}^{3}$ He concentration allowed us to characterize nearly all the static and dynamic properties of the dislocation network. Such a set of results and the way how they were obtained is clearly particular to this quantum crystal. There are a few more questions which would deserve more study. One of them concerns the exact value of the kink energy: is it zero or not? A related one is the value of the Peierl's barrier for the motion of existing kinks: once more is it zero or not?

It would be nice also to determine the width of split dislocation. It is related to the energy of stacking faults which is not yet precisely known.

The case of pure <sup>3</sup>He crystals is obviously interesting. They exist in two different structures: body centered or hexagonal compact. Dislocation properties should be rather different in the two structures. The tunneling of kinks and of impurities should be rather different from the <sup>4</sup>He case because <sup>3</sup>He atoms have a non-zero nuclear spin that is disordered except below 1 mK, an ultralow temperature that would make the study of mechanical properties very difficult. Furthermore, <sup>3</sup>He crystals are much more difficult to purify than <sup>4</sup>He ones.

Eventually, there remains some controversy about a possible mass flow along the dislocation cores. Experiments at the Massachusetts University (Amherst) [31] and in Edmonton (Canada) [32] are in progress, whose sets of results are not yet fully understood.

#### Acknowledgements

We acknowledge support from NSERC (Canada) and from the European Research Council grant AdG 247258-SUPERSOLID.

#### References

- [1] R.A. Aziz, A.R. Janzen, M.R. Moldover, Phys. Rev. Lett. 74 (1995) 1586.
- [2] F. London, Proc. R. Soc. A 153 (1936) 576.
- [3] E.J. Rugeles, S. Ujevic, S.A. Vitiello, arXiv:1503.1163 [cond-mat.other], 3 March 2015.
- [4] F. Souris, A.D. Fefferman, H.J. Maris, V. Dauvois, P. Jean-Baptiste, J.R. Beamish, S. Balibar, Phys. Rev. B 90 (2014) 180103.
- [5] C. Pantalei, X. Rojas, D.O. Edwards, H.J. Maris, S. Balibar, J. Low Temp. Phys. 159 (2010) 452.
- [6] F.A. Lindemann, Phys. Z. Sowjetunion 11 (1910) 609.
- [7] C.A. Burns, E.D. Isaacs, Phys. Rev. B 55 (1997) 5767.
- [8] For a review, see S. Balibar, H. Alles, A.Ya. Parshin, Rev. Mod. Phys. 77 (2005) 317.
- [9] For some more details on the observation of helium crystals, see also S. Sasaki, F. Caupin, S. Balibar, J. Low Temp. Phys. 153 (2008) 43.
- [10] E. Kim, M.H.W. Chan, Nature 427 (2004) 225.
- [11] E. Kim, M.H.W. Chan, Science 305 (2004) 1941.
- [12] J. Day, J. Beamish, Nature 450 (2007) 853.
- [13] J.R. Beamish, A.D. Fefferman, A. Haziot, X. Rojas, S. Balibar, Phys. Rev. B 85 (2012) 180501.
- [14] H.J. Maris, Phys. Rev. B 86 (2012) 020502.
- [15] D.Y. Kim, M.H.W. Chan, Phys. Rev. Lett. 109 (2012) 155301.
- [16] A. Haziot, X. Rojas, A.D. Fefferman, J.R. Beamish, S. Balibar, Phys. Rev. Lett. 110 (2013) 035301, and its supplemental material.
- [17] J. Friedel, Dislocations, Pergamon, New York, 1964.
- [18] J. Friedel, private communication to S. Balibar, 2013.
- [19] B. Legrand, Philos. Mag. 49 (1984) 171.
- [20] F. Souris, A.D. Fefferman, A. Haziot, N. Garroum, J.R. Beamish, S. Balibar, J. Low Temp. Phys. 178 (2015) 149.
- [21] A. Haziot, X. Rojas, A.D. Fefferman, J.R. Beamish, S. Balibar, Phys. Rev. Lett. 111 (2013) 119601.
- [22] D.O. Edwards, S. Balibar, Phys. Rev. B 39 (1989) 4083.
- [23] A. Granato, K. Lucke, J. Appl. Phys. 27 (1956) 583.
- [24] T. Ninomiya, J. Phys. Soc. Jpn. 36 (1974) 399;
- T. Ninomiya, Scr. Metall. Mater. 18 (1984) 669.
- [25] A.D. Fefferman, F. Souris, A. Haziot, J.R. Beamish, S. Balibar, Phys. Rev. B 89 (2014) 014105.
- [26] A. Haziot, A.D. Fefferman, F. Souris, J.R. Beamish, H.J. Maris, S. Balibar, Phys. Rev. B 88 (2013) 014106.
- [27] J. Friedel, C. Boulanger, C. Crussard, Acta Metall. 3 (1955) 380.
- [28] A. Allen, M. Richards, J. Schratter, J. Low Temp. Phys. 47 (1982) 289.
- [29] N. Sullivan, Appl. Magn. Reson. 8 (1995) 361.
- [30] S.S. Kim, C. Huan, L. Yin, J.S. Xia, D. Candela, N. Sullivan, Phys. Rev. B 87 (2013) 224303.
- [31] See a series of articles from M.W. Ray, R.B. Hallock, Phys. Rev. Lett. 100 (2008) 235301; to Ye. Vekhov, W.J. Mullin, R.B. Hallock, Phys. Rev. Lett. 115 (2015) 019902.

- [32] Z.G. Cheng, J. Beamish, A.D. Fefferman, F. Souris, S. Balibar, V. Dauvois, Phys. Rev. Lett. 114 (2015) 165301.
  [33] W.L. Bragg, Proc. Phys. Soc. 52 (1940) 54.
  [34] E.R. Parker, J. Washburn, Trans. Metall. Soc. AIME 194 (1952) 1076.
  [35] A. Haziot, A.D. Fefferman, J.R. Beamish, S. Balibar, Phys. Rev. B 87 (2013) 060509.