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Re-entrant peak effect in an anisotropic superconductor $2H\text{-}NbSe_2$: Role of disorder

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Abstract. – The influence of disorder (induced by pinning centres) on the peak effect (PE) phenomenon, namely, an anomalous increase in J_c *vs.* H (or T), has been studied in the anisotropic superconductor $2H\text{-}NbSe_2$. Results from new electrical transport as well as dc and ac magnetic response studies are presented to demonstrate that increasing disorder shrinks the (H, T) parameter space over which the vortex lattice retains spatial order. We find that the re-entrant nature of PE is clearly manifested only in crystals which have moderate amount of disorder. We also find that the upper branch of the PE curve is fairly robust, whereas the lower re-entrant branch is strongly affected by disorder.

The advent of high-temperature superconductors (HTSC) focused attention on the influence of thermal fluctuations on Abrikosov flux line lattice (FLL) or vortex lattice (VL) and led to the prediction [1] of a *vortex liquid state* with an unusual re-entrant melting (H_m, T_m) phase boundary in the field-temperature (H, T) plane such that at a fixed T, the phase boundary $H_m(T)$ is encountered *twice* upon increasing H [2]. A dilute vortex liquid phase is expected to form just above the lower critical field H_{c1}, where the lattice constant $a_0(\propto \frac{1}{\sqrt{\text{Field}}}) \geq \lambda$, and a highly dense vortex liquid phase is expected below the upper critical field H_{c2}, where $a_0 \geq 2\xi$. The two branches comprising the re-entrant melting phase boundary join around the so-called "nose" temperature above which the vortex array is thermally melted at all field values. The

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upper branch of the melting curve is given approximately by a quadratic Lindemann variation, $\beta_m\,(c_L^4/G_i)H_{c2}(0)(1-T/T_c)^2$ (see eq. (5.5) of ref. [2]), where c_L is the Lindemann parameter, G_i is the Ginzburg number ($= (1/2)(k_B T_c/H_c^2\xi^3\epsilon/8\pi)^{1/2}$) and $\beta_m \approx 5.6$. G_i is much larger in the cuprate HTSC systems as compared to that in low T_c alloys, which facilitates the observation of the dense vortex liquid phase over an appreciable (H, T) region below the $H_{c2}(T)$ line in HTSC systems [2]. However, the large value of κ and the small value of H_{c1} in the cuprates presumably [2] restricts the dilute vortex liquid phase to very small induction values, making its experimental observation a challenging task. A further difficulty in the observation of the dilute vortex liquid phase arises due to the ubiquitous pinning centers in all real systems which adversely affects the translational symmetry and the stability of the ordered VL. The effect of quenched disorder is expected to be especially strong for dilute vortex arrays and may mask the observation of an underlying dilute liquid-dilute solid freezing transition [3,4]. In the context of quenched disorder, the phenomenon of the peak effect (PE), an anomalous increase in the critical current, is continuing to receive a great deal of attention [5-12]. In a collective pinning description [13], J_c relates inversely to the volume V_c of the Larkin domain ($J_c \propto \frac{1}{\sqrt{V_c}}$) over which FLL is correlated. The anomalous peaking in J_c indicates a rapid decrease in V_c due to softening of the elastic moduli of FLL at the incipient melting transition [5,10,11,13].

Ghosh *et al.* [9] showed that in a weakly pinned crystal of hexagonal 2H-$NbSe_2$ ($T_c \approx 7.1$ K), the PE curve, which is the locus of peak temperatures $T_p(H)$ (determined from the magnetic shielding response, *i.e.* ac susceptibility χ' measurements), bore a striking resemblance to the theoretically proposed re-entrant [1,2] melting phase boundary $T_m(H)$. 2H-$NbSe_2$ has an appreciable value ($\approx 3\times10^{-4}$) of G_i, which lies between those of HTSC and conventional superconducting alloys [10]. The *turnaround* of the re-entrant $T_p(H)$ curve in 2H-$NbSe_2$ has been reported [9] to occur when a_0(of 4000 Å at $H \approx 150$ Oe) $\approx \lambda_c$ (in 2H-$NbSe_2$), and it is followed by a rapid broadening of PE. In this letter, we report new results on the re-entrant nature of the PE curve and the effect of quenched random disorder on it via electrical transport measurements and an equilibrium dc magnetization experiment on the specimen of 2H-$NbSe_2$ utilized by Ghosh *et al.* [9], and magnetic shielding (χ') response studies on two other different crystals of the same compound with qualitatively different levels of quenched random disorder. The conventional dc electrical transport results not only fortify the earlier claim [9] of the re-entrant nature of the $T_p(H)$ curve but also corroborate the previous finding that PE broadens across fields where *turnaround* in the $T_p(H)$ curve takes place. The magnetization hysteresis data confirm the double crossover of the $T_p(H)$ curve in an isothermal scan near the *turnaround* region. We further find that though the upper (higher H) branch of the $T_p(H)$ curve is somewhat robust, the lower re-entrant branch is strongly influenced by disorder. Decreasing pinning strength results in the "nose" region being pushed down to fields lower than that reported earlier [9], whereas increasing pinning could make the "nose" feature completely disappear.

Three single crystals of 2H-$NbSe_2$ with increasing quenched disorder used in the present study comprise platelets belonging to the same batches as the specimen X used by Higgins and Bhattacharya [10], as the specimen Y used by Ghosh *et al.* [9] and as the specimen Z used by Henderson *et al.* [12]. The temperature-dependent dc resistance $R(t)$ data to ascertain PE temperatures in specimen Y were recorded following ref. [10], the isothermal magnetization data to identify the manifestation of PE in a similar sample were obtained on a Quantum Design Inc. Squid magnetometer following *the half scan technique* of Ravikumar *et al.* [14], and ac susceptibility data in all three samples were recorded using a high-sensitivity ac susceptometer [9]. The essential new findings are summarized in figs. 1 to 3.

Figure 1 shows R *vs.* reduced temperature ($t = T/T_c(0)$) in the crystal Y at some of the low fields (120 Oe to 380 Oe) to elucidate the re-entrant behavior in $t_p(H)$ data. The inset (a) of

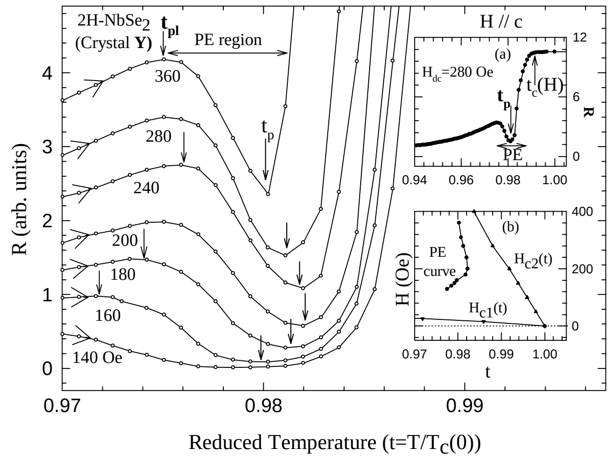

Fig. 1. – The inset panel (a) shows resistance *vs.* reduced temperature ($t = T/T_{\mathrm{c}}(0)$) in $H_{\mathrm{dc}} = 280$ Oe ($\parallel c$) in the crystal Y of 2H-NbSe$_2$. The main panel shows portions of the R *vs.* t curves in the PE regions at some chosen dc fields. The inset panel (b) shows the re-entrant nature of $t_{\mathrm{p}}(H)$.

fig. 1 shows that the peak in J_{c} at a given H manifests itself as an anomalous dip in $R(t)$. As emphasized in ref. [10], the location of each $t_{\mathrm{p}}(H)$ is robust. The main panel of fig. 1 shows portions of the $R(t)$ curves and focuses attention on the identification of $t_{\mathrm{p}}(H)$ values. The inset panel (b) of fig. 1 depicts the PE curve passing through $t_{\mathrm{p}}(H)$ for $120 \leq H \leq 380$ Oe. The shape of the PE curve clearly demonstrates its re-entrant characteristics, with *turnaround* occurring at about 180 Oe, in excellent agreement with earlier results [9]. The anomalous PE behavior in a given $R(t)$ curve starts at a temperature t_{pl}, where the resistance starts to decrease (see the t_{pl} mark for the $H = 360$ Oe curve). The significant broadening of the PE region starts to occur as the H values are decreased below 200 Oe, and eventually below 120 Oe, the PE becomes so shallow that $t_{\mathrm{p}}(H)$ cannot be located precisely.

The inset panel of fig. 2 shows a portion of the dc M-H hysteresis data at $T = 6.95$ K in crystal Y of Ghosh *et al.* [10]. In an isothermal scan at $T = 6.95$ K, we expect to encounter both the lower and upper branches of the re-entrant PE curve. The PE on the upper branch of $T_{\mathrm{p}}(H)$ curve manifests itself as an anomalous opening of the hysteresis loop before reaching the H_{c_2} value. According to Bean's Critical State Model [15], the magnetization hysteresis $\Delta M(H)$ (difference between forward and reverse magnetization values at given H) is a measure of $J_{\mathrm{c}}(H)$. To locate the existence of PE on the lower branch and to elucidate that the peak field H_{p} on this branch *increases* as T *increases*, we show $4\pi\Delta M(\propto J_{\mathrm{c}}(H))$ *vs.* H at 6.9 K, 6.95 K and 7.0 K in the low-field region in fig. 2. Note that for $H < 60$ Oe, the $\Delta M(H)$ values at 6.95 K are *lower* than those at 6.9 K and this reflects the normal behavior in $J_{\mathrm{c}}(H,T)$. However, a crossover in the $\Delta M(H)$ curves occurs at about 70 Oe such that the $\Delta M(H)$ values at 6.95 K *exceed* those at 6.9 K, thereby exemplifying anomalous behavior [16] in $J_{\mathrm{c}}(H,T)$ due to PE on the lower branch of the $T_{\mathrm{p}}(H)$ curve at 6.95 K. Further note that the $\Delta M(H)$ values at 7.0 K become larger than those at 6.95 K above 130 Oe. Thus, at 7.0 K the (lower) peak effect is seen to be encountered at a field value larger than that at 6.95 K.

Figures 3(a) to 3(c) show the in-phase ac susceptibility (χ') *vs.* reduced temperature t at 211 Hz and in h_{ac} of 1 Oe (r.m.s.) at some of the chosen dc fields H_{dc} (0 to 2 kOe) in three crystals X, Y and Z of 2H-NbSe$_2$. These crystals were grown by the same vapor transport method [10], but with starting materials of different purity [9, 10, 12]. The relative purity

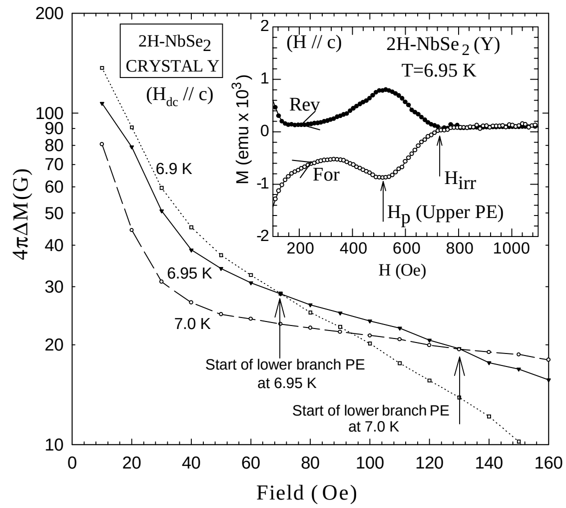

Fig. 2. – Isothermal magnetization hysterisis data $[4\pi\Delta M = 4\pi(M_R - M_F)]$ for $H_{\rm dc} \parallel c$ in the crystal Y of 2H-NbSe$_2$ at the temperatures indicated. The inset panel shows a portion of the M-H loop showing upper PE at 6.95 K.

of the three crystals is reflected in the progressively enhancing *width* of the superconducting transition in zero field. In a $\chi'(t)$ measurement, the PE is identified by an anomalous negative peak [8,9]. The peak temperatures $T_{\rm p}(H)$ are independent of frequency and amplitude of the ac field $h_{\rm ac}$ in the parametric range of our experiment [17]. Figure 3(a) shows that in the most weakly pinned crystal X, the PE peaks are very sharp; *their half widths are smaller than the width of superconducting transition in zero field.* This fact supports the association of $T_{\rm p}(H)$ with a possible phase transition, like the phenomenon of change in spatial order of the vortex array across $T_{\rm p}$ [11].

Figure 3(b) is based on the $\chi'(t)$ response in crystal Y [9]. $T_{\rm p}(H)$ values for $H = 300$ Oe and $H = 100$ Oe can be seen to be nearly the same, consistent with the re-entrant nature of the PE curve in crystal Y [9]. However, the PE in $H = 200$ Oe (data not shown) is significantly broader than that at $H = 300$ Oe. Figure 3(c) shows the $\chi'(t)$ response in the most strongly pinned [13] crystal Z. In this sample, the PE peak is considerably broad even at $H = 1000$ Oe as compared to the corresponding peaks in crystals X and Y (cf. figs. 3(a) to (c)). The PE peak is barely discernible at $H = 300$ Oe in crystal Z and below 200 Oe, the PE manifests only as a hump/kink in the $\chi'(t)$ curve.

Figure 4 summarizes the $T_{\rm p}(H)$ data in crystals X, Y and Z as H *vs.* $T_{\rm p}/T_{\rm c}(0)$ curves. For the sake of convenience and reference, this figure includes the $H_{{\rm c}_1}(t)$ and $H_{\rm c2}(t)$ curves obtained in crystal X [9]. Thus, from figs. 3 and 4, it is apparent that the enhanced quenched random disorder (pinning) results in the following effects: (a) For a given $H(H \geq 300$ Oe), the PE peak occurs at a lower value of t (see, for instance, arrows marking $T_{\rm p}(H)$ values in three crystals at $H = 300$ Oe in figs. 3(a) to 3(c)). (b) The re-entrant lower branch of the PE curve is clearly evident only for crystal Y with intermediate level of disorder and not so for the other two crystals. The PE curve in crystal X shows a steep fall for $200 < H < 30$ Oe and its turnaround characteristic presumably lies much below 30 Oe; however, PE in $\chi'(T)$ data is unobservable in crystal X for $H \leq 20$ Oe. In sample Z, the PE becomes so broad at 300 Oe that the precise location of $T_{\rm p}(H)$ below this field becomes somewhat ambiguous (see fig. 3(c)). However, it can be safely surmised that the PE curve at lower fields ($H < 500$ Oe) moves away from the values that can be extrapolated from the $T_{\rm p}(H)$ *vs.* H data at higher

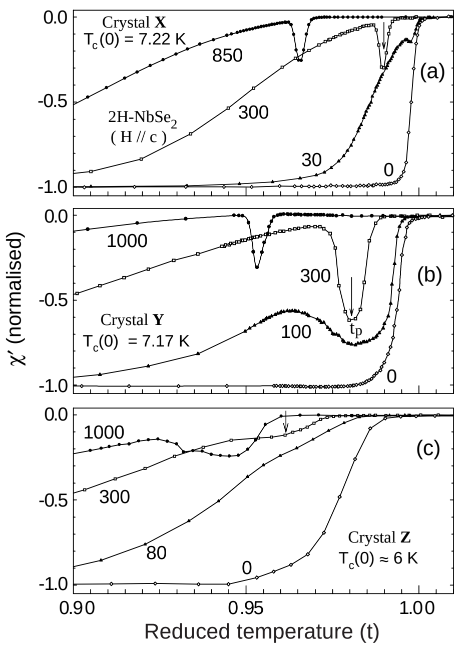

Fig. 3

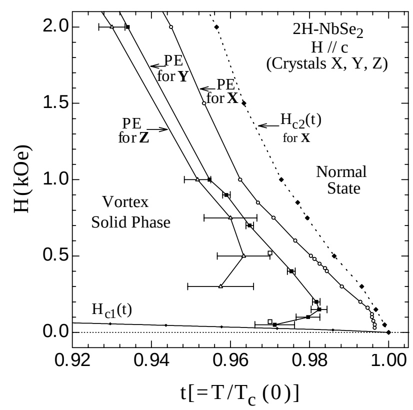

Fig. 4

Fig. 3. – In-phase ac susceptibility (χ') ($\omega = 211$ Hz, $h_{\rm ac} = 1$ Oe (r.m.s.)) *vs.* t $(= T/T_{\rm c}(0))$ in crystals X, Y and Z in fixed dc fields ($\parallel c$).

Fig. 4. – Magnetic phase diagram in three crystals of 2H-NbSe$_2$. The PE curves and $H_{\rm c2}(T)$ curve correspond to the $T_{\rm p}(H)/T_{\rm c}(0)$ and $T_{\rm c}(H)/T_{\rm c}(0)$ values obtained from $\chi'(t)$ data as in fig. 3. For crystal Y, the two open squares lying on the upper and lower branches of the PE curve identify the peak field ($H_{\rm p}$) and crossover field at 6.95 K as in fig. 2.

fields ($H > 500$ Oe).

The results of figs. 1, 3 and 4 provide the following general conclusions:
i) For the upper branch (where $\mathrm{d}T_{\rm p}/\mathrm{d}H < 0$) of the PE curve, increasing pinning reduces the volume $V_{\rm c}$ of the Larkin domain, which then requires less thermal fluctuations to melt/amorphize it around $t_{\rm p}$. The quenched disorder and thermal fluctuations conspire together to destabilize the ordered phase and a "lowering" of the $t_{\rm p}(H)$ curve is seen. ii) For the lower branch (where $\mathrm{d}T_{\rm p}/\mathrm{d}H > 0$), increasing pinning reduces $V_{\rm c}$ as before. Thus, one needs enhanced interaction (*i.e.* increase in H) to stabilize the ordered phase and the "raising" of the PE curve occurs. iii) Increasing pinning has the general effect of masking the difference between a vortex solid and a vortex liquid, converting both into a "glassy" state. A clear distinction between the two phases is no longer possible, the transformation process is more gradual and thus a broadening of the PE results. iv) As can be ascertained from the main panel of fig. 1, the onset (at $t_{\rm pl}$) of PE, which is apparently driven by quenched disorder, is also re-entrant. Recent muon spin rotation studies [18] on crystals of 2H-NbSe$_2$ have provided microscopic evidence in favor of a sudden change in spatial order of FLL at $t_{\rm pl}$.

Finally, it is of interest to examine the scenario emerging from fig. 4 with the expectations of theoretical studies. In the cleanest crystal X, where we observe only the upper branch of the PE curve, which most closely marks the transformation of the ordered solid into the *pinned "liquid" state* [10] and a narrow region separates this line from the $T_{\rm c}(H)$ line (cf. figs. 6, 24 and 25 of ref. [2] and our fig. 4). In between $T_\mathrm{p}(H)/H_\mathrm{p}(T)$ and $T_\mathrm{c}(H)/H_{\mathrm{c}_2}(T)$ lines, the pinned liquid becomes unpinned at $H_\mathrm{irr}(T)$ (see fig. 2(a) for location of H_irr at 6.95 K). Fitting the higher field ($H > 200$ Oe) branch of the PE curve to $\beta_\mathrm{m}(c_\mathrm{L}^4/G_i)H_{\mathrm{c}2}(0)(1-T/T_\mathrm{c})^2$ with $G_i \sim 3\times10^{-4}$ and $H_{\mathrm{c}_2}(0) \approx 4.6$ T [10], we get a reasonable value for $c_\mathrm{L} = 0.15$. A pronounced departure (in $T_\mathrm{p}(H)$ at $H \leq 200$ Oe) away from the upper portion appears in accord with the work of Blatter and Geshkenbein [19], who found a similar rapid drop in the FLL melting curve (in the absence of pinning effects) away from the H_{c_2} phase boundary at low fields (*i.e.* above the *turnaround* feature of the melting boundary). The re-entrant lower branch of the PE curve may thus be at still lower fields in crystal X, as proposed theoretically [19], and outside the detection capability of the present measurements. In crystal Y, Ghosh *et al.* [9] estimated c_L as about 0.17 from higher H portion of the $T_\mathrm{p}(H)$ data. They also showed that if the Nelson-Le Doussal line [3] indeed marks the *crossover* from an interaction-dominated to a disorder-dominated region, then the ratio of entanglement length L_E to the pinning length L_c becomes approximately equal to 1 ($L_\mathrm{c}/L_\mathrm{E} \sim 1$) at $B = 30$ Oe and $t = 0.975$. $L_\mathrm{c}/L_\mathrm{E}$ is given as [2]

$$\frac{L_\mathrm{c}}{L_\mathrm{E}} = \left(\frac{\pi\kappa^2\ln(\kappa)}{\sqrt{2}}\right)\left(\frac{a_0^2}{2\pi\lambda(0)}\right)\frac{(J_\mathrm{c})^{1/2}}{(G_i j_o)^{1/2}}\frac{(1-t)^{4/3}}{t}\,, \tag{1}$$

where various symbols have their usual meaning (see eq. (6.47) of ref. [2]). Below a crossover field of 30 Oe, the vortex array would be in disentangled liquid or glass state [2-4]. Equation (1) implies that the field at which crossover to glassy state can occur varies as $J_\mathrm{c}^{1/2}$. As compared to crystal Y, J_c is larger in crystal Z by nearly a factor of 50. This, coupled with the observation that in crystal Z the PE becomes difficult to discern below $t \approx 0.96$, yields a value of crossover field of $\sim$ 400 Oe, which demonstrates a reasonable agreement with an observed value of 300 Oe.

In summary, we have examined how a variation in the disorder (and hence pinning) influences the PE in 2H-NbSe$_2$. We have shown that with increasing disorder, the PE broadens and shifts in the (H, T) space such that the ordered lattice is destabilized on both the upper and lower brances of the PE "phase boundary". As a result of these shifts at high temperatures and low fields, the lower re-entrant branch can be easily observed experimentally only in crystals with moderate disorder. Some aspects of our results can also be semi-quantitatively analysed using currently available theories [3, 4, 19, 20].

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