Reentrant Peak Effect and Melting of a Flux Line Lattice in 2H-$NbSe_2$

K. Ghosh, S. Ramakrishnan,* A. K. Grover,* Gautam I. Menon, and Girish Chandra
Tata Institute of Fundamental Research, Bombay 400005, India

T. V. Chandrasekhar Rao, G. Ravikumar, P. K. Mishra, and V. C. Sahni
Solid State Physics Division, Bhabha Atomic Research Centre, Bombay 400085, India

C. V. Tomy, G. Balakrishnan, and D. Mck Paul
Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom

S. Bhattacharya
NEC Research Institute, 4 Independence Way, Princeton, New Jersey 08540
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A reentrant peak effect is observed through low field ac susceptibility measurements on the weakly pinned flux line lattice in single crystals of 2H-$NbSe_2$. The resulting phase diagram of the peak effect is strikingly similar to the theoretically predicted reentrant phase boundary which separates flux lattice and flux liquid phases. The broadening and ultimate disappearance of the peak effect at very low fields is consistent with the predicted crossover to a disordered glassy state in this field regime. [S0031-9007(96)00374-2]

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The melting transition of the flux line lattice (FLL) in type-II superconductors has been a subject of intense experimental and theoretical study since the first experiments on the irreversible-reversible transition by Mueller *et al.* [1]. Much of this effort has focused on the high T_c cuprates in which elevated transition temperatures, large penetration depths, and considerable anisotropy conspire to enhance the (H, T) regime where a thermally melted flux liquid phase occurs [2–6]. A particularly interesting prediction is that of the existence of a reentrant behavior of the melting phase boundary at low fields, first proposed by Nelson [2] and further elaborated in numerous other theoretical studies (see, for instance, Figs. 2, 3, and 6 in [5]). At low field values, the melting temperature $T_M(H)$ is expected to increase with increasing field, reflecting the gradual stiffening of the lattice. This phase boundary should cross over to the more commonly studied high field regime [7,8], where $T_M(H)$ *decreases* with increasing H. The low field flux liquid region sandwiched between Meissner and vortex solid phases had, however, remained elusive so far.

The striking phenomenon of an abrupt and nonmonotonic increase of the critical current slightly below H_{c2}, commonly known as the "peak effect" (PE), has also received a great deal of attention in recent years [9–11]. It has been argued in several contexts [10–12] that the PE is closely related to the underlying melting of the FLL, in an extension of the original Pippard [13] scenario of the PE which suggested that the reduction of the shear modulus close to the upper critical field could occur faster than the reduction of the pinning force of the inhomogeneities, resulting in an anomalous increase in the critical current density. Even in situations where the PE is considered to occur only slightly below [14] the melting transition, the temperature variation of the PE is believed to track the melting transition extremely closely. Moreover, several studies have shown that its locus is as robust [9–11] as the critical fields themselves are and can often be located with greater ease.

In this Letter we report the experimental observation of a novel feature involving the peak effect phenomenon in a weakly pinned flux line lattice: At low fields the peak effect shows a reentrance behavior, yielding a locus remarkably similar to the theoretically expected reentrant melting phase boundary. The experiments involving ac susceptibility measurements were performed on single crystal samples (grown by vapor transport method [15]) of the layered anisotropic superconductor 2H-$NbSe_2$ ($\gamma \approx 3$, $T_c \approx 7.1$ K) [16], which has recently received much attention due to its extremely low critical current densities and the ubiquitous PE in nonlinear transport and magnetic measurements slightly below the upper critical fields [9–11]. The PE temperatures $T_{\rm peak}(H)$ in the present study are identified with the positions of negative peaks (i.e., maxima in diamagnetic screening responses as a consequence of peaking of the critical current density) in the in-phase ac susceptibility recorded in the presence of superposed dc fields at low frequencies (<1 kHz) [17]. Figures 1(a) and 1(b) show plots of $\chi'_H(T)$, for several $H(\|c)$ values, recorded at a frequency of 21 Hz with an ac amplitude of 2 Oe (rms) in a representative single crystal of 2H-$NbSe_2$. In a nominal zero field, the diamagnetic screening response is seen to monotonically decrease [see Fig. 1(a)] as T tends to $T_c(0)$ (i.e., reduced temperature $t \rightarrow 1$). However, for $100 < H < 1000$ Oe, each of the $\chi'_H(T)$ plots in Figs. 1(a) and 1(b) displays a clear negative peak [see also Fig. 4(a) of [18] and Fig. 2 of [9]] just

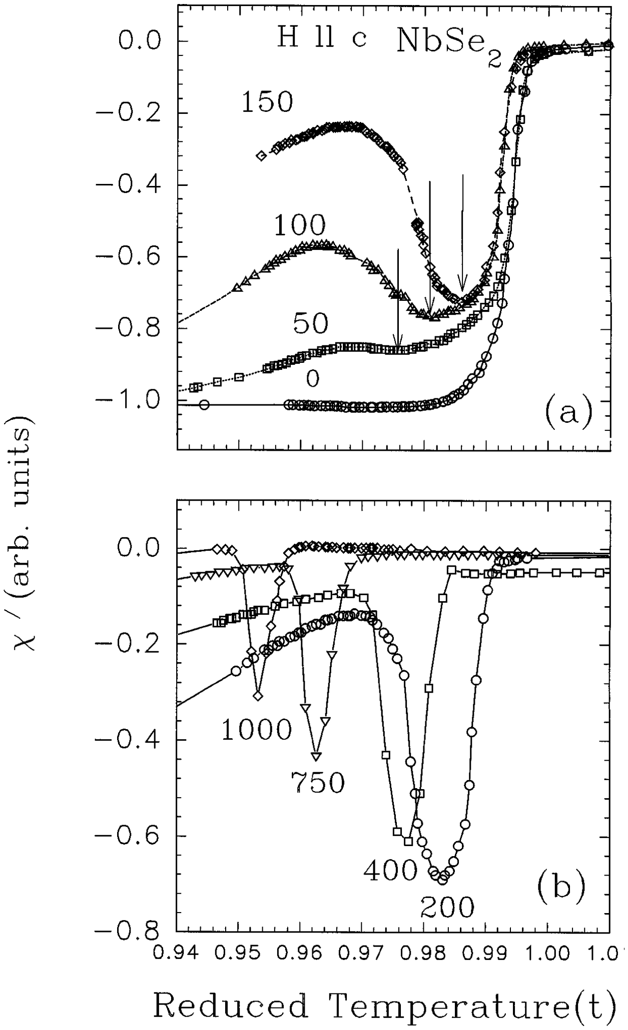

FIG. 1. Temperature variation of the in-phase ac [$f = 21$ Hz, $h_{ac} = 2$ Oe (rms)] susceptibility $\chi'_H(T)$ for $H \| c$ in a single crystal of 2H-NbSe$_2$ [$T_c(0 \approx 7.1$ K)]. The arrows identify the positions ($T_{\text{peak}}(H)$) of negative peak in $\chi''(T)$ plots.

before the diamagnetic screening response reaches the base line at $T_c(H)$. The arrows in Fig. 1(a) locate the $T_{\text{peak}}(H)$ values. As expected, $T_c(H)$ values monotonically increase as H decreases from 1000 Oe to zero field. $T_{\text{peak}}(H)$ values would appear to embrace the $T_c(H)$ [i.e., $H_{c2}(T)$] line from 1000 to 200 Oe [see Fig. 1(b)]. However, from 150 to 50 Oe [see Fig. 1(a)], $T_{\text{peak}}(H)$ values are observed to decrease instead of increasing [$dT_{\text{peak}}(H)/dH > 0$ for $H < 200$ Oe]. The change in sign of $dT_{\text{peak}}(H)/dH$ occurs between 200 and 100 Oe reproducibly and has been confirmed in a number of crystals of 2H-NbSe$_2$ for both $H \| a$ and $H \| c$ [19]. The data for $H \| a$ are not being shown here for brevity. Figure 2 shows the resulting magnetic phase diagram which comprises the $T_c(H)$ and $T_{\text{peak}}(H)$ data for $H \| c$ in the 2H-NbSe$_2$ system. The phase diagram also contains the values of the lower critical field for $H \| c\,[H_{c1}(T)]$ [20]. Note the remarkable similarity between the $T_{\text{peak}}(H)$ line in Fig. 2 and the theoretically predicted melting line(s) [2,5]. This similarity provides justification for earlier speculations that the $T_{\text{peak}}(H)$ line in a weakly disordered system closely approximates the melting line of the disorder-free FLL. In what follows we examine our results in light of available theories on FLL melting in order to check if such an aformentioned similarity between the two phase boundaries withstands more rigorous quantitative tests.

We show in the inset of Fig. 2 the plot of $[1 - T_{\text{peak}}/T_c(0)]^2$ vs $H(\|c)$. The upper portions of the $T_{\text{peak}}(H)$ curve in Fig. 2 do conform to the relation $H \propto [1 - T_M(H)/T_c(0)]^\alpha$ [5] with $\alpha = 2$, as derived by Houghton *et al.* [3] and Blatter *et al.* [4]. From the slope value of the linear fit in the inset of Fig. 2, we estimate the Lindemann number c_L using the relation [5]

$$B_m(T) = \beta_m(c_L^4/\text{Gi})H_{c2}(0)(1 - T/T_c)^2, \qquad (1)$$

where $H_{c2}(0) \approx 4.6T$ [10], $\beta_m = 5.6$ [5], and $\text{Gi} \approx 3 \times 10^{-4}$, to be about 0.17. This value is in satisfactory agreement with all estimates obtained so far for Lindemann melting.

The turnaround of the $T_{\text{peak}}(H)$ curves between 200 and 100 Oe is a novel feature which implies that an isotherm (fixed T line) would intersect the PE curve twice. Across both these intersections, one anticipates manifestations of the PE phenomenon. Figure 3 shows isothermal χ' data for $H \| c$ in another crystal of 2H-NbSe$_2$. In this sample, the $T_{\text{peak}}(H)$ values at $H = 100$ and 200 Oe are nearly equal (data not shown here [19]) which confirms the multivalued characteristic of the PE curve. In Fig. 3, the diamagnetic response at a fixed t is seen to monotonically decrease with increase in H prior to the emergence of a *negative peak* while approaching $H_{c2}(T)$. The field value (H^u_{peak}) corresponding to the negative peak locates the

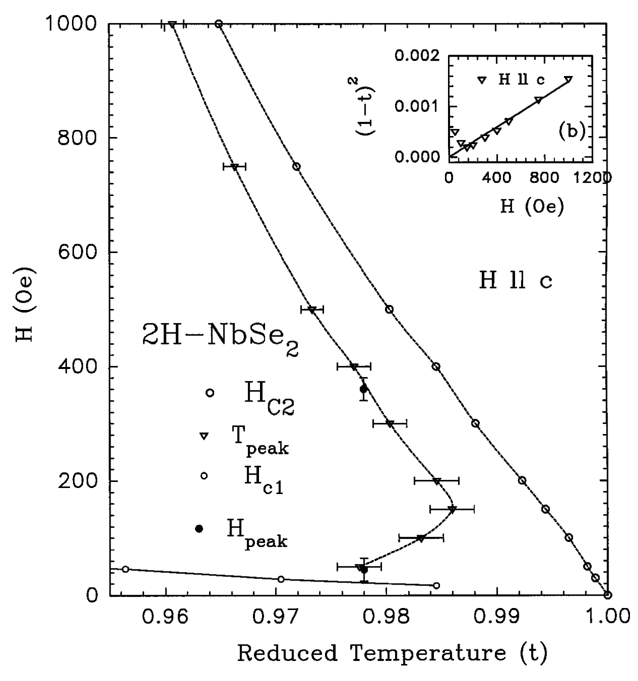

FIG. 2. Magnetic phase diagram near $T_c(0)$ in the 2H-NbSe$_2$ system. The inset shows the variation of $[1 - T_{\text{peak}}(H)/T_c(0)]^2$ with field for $H \| c$.

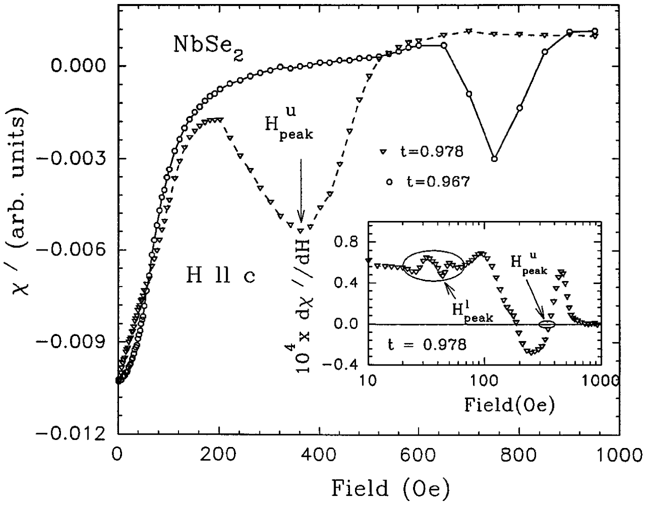

FIG. 3. Variation of χ' vs $H(\|c)$ at $t = 0.967$ and 0.978 in another single crystal of 2H-$NbSe_2$. The field region corresponding to negative peaks at $t = 0.967$ and 0.978 have been encircled in the main panel. The increase in diamagnetic response at $t = 0.978$ over that at 0.967 above 60 Oe is interpreted as the first intersection of the PE curve by the isotherm at $t = 0.978$. The second intersection of the same isotherm with the PE curve is shown by the negative peak around 380 Oe in the main panel. The inset shows $d\chi'/dH$ vs H at $t = 0.978$. The two intersections at $t = 0.978$ are located as $H^l_{\rm peak}$ and $H^u_{\rm peak}$. The occurrence of a double intersection of the PE curve by an isotherm in the reentrant region has been confirmed in dc magnetization experiments as well [21].

crossover of the PE curve at the upper field end in an isothermal run. Further, it may be noted that the isotherm at $t = 0.978$ (Fig. 3) crosses the isotherm at $t = 0.967$ at about 60 Oe. Below 60 Oe, the diamagnetic screening response at higher temperature value ($t = 0.978$) is smaller than that at lower temperature value ($t = 0.967$), which is the normal behavior of a superconductor. However, above 60 Oe, the inequality gets reversed [21]. The observation of a larger screening signal at a higher temperature is the manifestation of the PE phenomenon while crossing the $T_{\rm peak}(H)$ curve at the lower field end in an isothermal run. Another demonstration of the double PE at $t = 0.978$ is obtained by plotting $d\chi'/dH$ vs H as shown in the inset of Fig. 3 (this method of locating the lower field value of $H^l_{\rm peak}$ where PE occurs at $t = 0.978$ removes the effect of the large slope of the χ'-H curve in the low field region). Note the occurrence of two anomalies of nearly identical nature but of different magnitude, corresponding to the double occurrence [22] of PE as marked by encircling the field regions over which PE phenomenon occurs. The error bar on $H^l_{\rm peak}$ is larger; however, the upper field value $H^u_{\rm peak}$ can be precisely located. These two field values are shown as filled circles in Fig. 2. The near agreement of data extracted from isothermal run with those obtained from isofield runs (see Fig. 1) elucidates the robustness of the PE phenomenon.

We now turn to an analysis of the above results in the lower branch of the phase boundary. On dimensional grounds, the turnaround of the melting line (the "nose") should occur at field values for which the intervortex spacing, $a = \sqrt{2/\sqrt{3}}\,(\sqrt{\phi_0/B})$, is of order the London penetration depth. At the turnaround field ($B \approx 200$ Oe), we estimate $a \simeq 4000$ Å, comparable to London penetration depths at the turnaround temperature, again showing that the PE occurs in the right range in magnetic induction and is consistent with simulations. Note, however, that it occurs at a reduced temperature value of $t \approx 0.99$, much greater than in the cuprates due to the smallness of the Ginzburg number ($\mathrm{Gi} \approx 3 \times 10^{-4}$) in this system, once again consistent with theoretical expectations. The field dependence of the melting line in 2H-$NbSe_2$ is also weak: Over the field range 150 to 50 Oe, $T_M(H)$ changes by as little as 1.5%.

We estimate the collective pinning length L_c using $L_c \sim \xi(j_0/j_c)^{1/2}$ [5], where j_0 is the depairing critical current, j_c is the critical current density, and $\xi(T)$ is the coherence length. Taking $j_0/j_c \sim 10^5$ and $\xi(T) \sim 10^3$ Å, in the field and temperature regime of interest (close to but below the reentrant PE line), we estimate L_c to be ~ 10 μm. As L_c is within an order of magnitude of the typical sample dimension (of order 100 μm), we expect that the transition we see closely approximates the behavior of the clean FLL.

It is believed that the regime of reentrant flux liquid at *low fields* and at *low temperatures* is so small that it may be very difficult to access experimentally in the cuprates [5]. Interestingly, however, the much smaller value of the Ginzburg-Landau parameter κ (≈ 9 for $H\|c$) for the present system would allow us to access this lower sliver of flux liquid more easily. An estimate for the width of this flux liquid region follows from $\Delta H \sim H_{c1}/\ln\kappa$ as $0.5H_{c1}$ [2]; such a number is consistent with data in Fig. 2.

Our observation of the PE, which is a collective property of flux lines in an interaction dominated regime, for fields down to about 50 Oe implies that individual pinning, usually important in samples of high-T_c materials at such low fields, is not dominant in this field range in 2H-$NbSe_2$. However, for $H < 50$ Oe, the negative peak feature in χ' broadens such that no signature of PE is evident in χ' vs T plot at $H \approx 30$ Oe. Moreover, the signature of PE cannot be distinguished from the hysteresis loss peak (not shown here) in χ'' data. This suggests a crossover to the pinning dominated regime at $H \approx 30$ Oe [23]. This result can also be compared with theoretical expectations based upon the Nelson–Le Doussal [23] line which marks the crossover to a disorder dominated regime at low fields on the lower branch of the melting curve. Using the relation [Eq. (6.47) of Ref. [5]]

$$\frac{L_E}{L_c} \simeq \frac{\pi\kappa^2\ln\kappa}{\sqrt{2}}\left[\frac{a_0}{2\pi\lambda(0)}\right]^2\left[\frac{j_c}{G_i j_0}\right]^{1/2}\frac{(1-t)^{4/3}}{t}, \quad (2)$$

where L_c is the pinning length and L_E is the entanglement length, and substituting all the experimentally measured parameters in the above equation, we find $L_c/L_E \approx 1$ around $H = 30$ G and $t = 0.975$ (where the peak effect disappears). In other words, when the pinning length becomes comparable to the entanglement length, disorder prevails, melting itself ceases to be meaningful, and, remarkably, the peak effect is absent in our experiment.

In summary, we have demonstrated the occurrence of the reentrance in the peak effect at low fields. Given that the PE tracks the melting of the ideal FLL, we conclude that the above observations are indicative of the existence of a reentrant melting of the FLL in the 2H-$NbSe_2$ system. The location and properties of the experimental phase boundary are in adequate quantitative agreement with theoretical predictions for the FLL melting line. Furthermore, the broadening of the PE at low fields is consistent with theoretical calculations implying a crossover to a disorder dominated regime [23] and the concomitant reduction of the Larkin volume [5]. The PE thus becomes unobservable in the regime where disorder dominates over interactions, i.e., in a situation where the notion of melting itself is highly ambiguous.

Our results would suggest that a search for the melting phase boundary and its reentrance may be fruitful in other weak pinning low κ systems with reasonable values of Ginzburg parameters. We conclude with a word of caution. Despite several recent suggestions that the peak effect is indeed a signature of, or a close precursor to, the FLL melting, the exact correlation between the two is not yet on a firm theoretical footing. Our results lend strong support to the point of view that the two are intimately related. We hope these results would stimulate theoretical and experimental efforts aimed at resolving this important question.

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*To whom correspondence should be addressed.

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