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Evidence for possible interaction between CDW and magnetic vortices in 2H-NbSe₂: comparison with 2H-NbS₂

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Abstract - We have performed vibrating reed ($f \sim 1$ kHz) and ac susceptibility ($f \sim 800$ kHz, i.e. RF penetration depth) measurements on superconducting 2H-NbSe₂ and 2H-NbS₂ in magnetic fields. For NbSe₂, a low temperature dissipation peak is correlated with a dip in vibrating reed resonant frequency and peak in ac susceptibility, signifying softening of the flux line lattice; these features are not observed in NbS₂. We suggest that the softening of the FLL is due to an interaction with the CDW, which does not occur in NbS₂.

The discovery of superconductivity at elevated temperatures in the layered cuprates has spurred research on vortex pinning and dynamics in superconductors in general. Properties that were originally thought to be unique to extremely anisotropic or high temperature materials, such as the cuprates, were found to exist in other, more "conventional" superconductors. For example, even isotropic low temperature superconductors such as Nb [1] and NbTi [2] were observed to exhibit reversible magnetic behavior near T_C , due to the "free" motion of vortices, and 2H-NbSe₂ vibrating reeds [$T_C = 7.2$ K] were observed to exhibit secondary dissipation peaks, well below their superconducting transitions [1,3]. While such peaks were initially associated with flux line lattice (FLL) phase transitions, such as melting [4,5], in the cuprates, Brandt has suggested that they may occur in all type-II superconductors as a consequence of vortex diffusion along different directions [6]; for example, such has been suggested to occur in the organic superconductor (ET)₂Cu(NCS)₂ [7].

The resonant frequency, f , of a vibrating superconducting reed in an external magnetic field H_0 increases when the FLL becomes pinned; for rigid pinning, Δf is proportional to the FLL tilt modulus, C_{44} [6,8], although screening currents greatly enhance the frequency shift by a geometric factor for $H_0 \perp \hat{c}$ [8]. ($\hat{c}$ is perpendicular to the conducting layers and parallel to the vibration direction.) For nonrigid pinning, Δf will be slightly less: $\Delta f_{\text{pinned}} - \Delta f \propto 1/\alpha$, where the Labusch parameter α is the curvature of the FLL pinning potential [8]. A dissipation peak reflecting the slowing of vortex motion with decreasing temperature accompanies the increase in f [6,8]. (For a vibrating reed, the dissipation is measured by the damping of the reed, $1/Q$, where Q is the quality factor.)

Two dissipation peaks are observed for NbSe₂ reeds; the lower temperature dissipation peak seems to be closely associated to an unusual dip in resonant frequency [3]. Both features are very sample and history dependent; the hysteretic behavior is inconsistent with the diffusion model [3], in which the temperature of the lower peak should depend only on the frequency, dimensions, and diffusion constant. Because of hysteresis, all data discussed in this paper will be for field cooling from the normal state. The lower dissipation peak and frequency dip are strongest for reeds oriented transverse to the external field, i.e. $\hat{c} \parallel H_0$ [3]. Typical vibrating reed results, with $\hat{c} \parallel H_0 = 0.73T$, are shown in Figure 1. The upper dissipation peak coincides with the rapid increase in resonant frequency and the onset of zero resistance (not shown) [3]. In Figure 2 are plotted H_0 vs. T for the upper dissipation peak (T_U), dip in frequency, T_F , and lower dissipation peak, T_L , for $\hat{c} \parallel H_0$. The dip in frequency, i.e., softening of the FLL, is almost unique to this material; softening at a much lower frequency (<1 Hz) has been observed in YBa₂Cu₃O₇, and associated with FLL melting [9].

Since the vibrating reed results are associated with motion of the FLL, related effects are expected to be observed in purely magnetic measurements. We have measured changes in the RF penetration depth for ac ($H_{ac} < 1$ Oe) fields parallel to $\hat{c}$ by measuring changes in the resonant frequency of an 800 kHz tank circuit inductively coupled to the crystal. The change in resonant frequency of the tank circuit is proportional to (minus) the change in penetration depth and therefore (minus) the change in effective susceptibility, χ' . Indeed, non-monotonic behavior of χ' as a function of temperature is observed. Results for $H_0 = 0.92T$ at different crystal orientations (θ is the angle between H_0 and the a,b plane) are shown in Figure 3. Results for, $\hat{c} \parallel H_0$, i.e. 90°, results are sensitive to compression of the FLL, i.e. C_{11} [10], while

Fig.1 - Resonant frequency shift and damping ($1/Q$) of a NbSe₂ vibrating reed ($f \sim 1.3$ kHz) vs. temperature. Curves are guides to the eye.

Fig.2 - Applied magnetic field vs. temperature for two crystals of NbSe₂ for various features of vibrating reed and ac susceptibility measurements described in text; lines through T_{ON} and $T_F \approx T_X$ are guides to the eye.

$\hat{c} \perp H_0$, 0° , results are sensitive to the FLL tilt modulus C_{44} . For $\hat{c} \parallel H_0$, the temperature of the onset of the frequency shift, T_{ON} , coincides closely with the center of the resistive transition, while the peak in χ' , T_χ , coincides with T_F , the dip in the vibrating reed frequency, as shown in Figure 2. Thus the magnetic and vibrating reed results indicate that C_{11} and C_{44} soften at the same temperature, while the magnitude of the softening decreases as the FLL becomes parallel to the ab plane and van der Waals gap. The three order of magnitude difference in frequency for the two measurements argues strongly against a simple diffusion mechanism for this feature.

For temperatures below T_U , the dissipation is hysteretic and strongly amplitude dependent [3], reflecting irreversible jumping of vortices [8]. We have started investigating the amplitude dependence of the magnitude of the frequency dips in both the magnetic and vibrating reed measurements. In our preliminary investigation, there does not seem to be a significant amplitude dependence for $\hat{c} \parallel H_0$, suggesting that in this orientation the softening is "elastic", i.e. due to a decrease in Labusch parameter.

Because the second dissipation peak is also seen in the cuprates, it was of interest to examine $2H-NbS_2$, which has a similar T_C (6.5K) but is more anisotropic than the selenide[11,12]. For example, the anisotropy in dH/dT_{ON} (as $H \rightarrow 0$), determined from the RF penetration depth, for both materials is shown

Fig.3 - Effective ac susceptibility for $NbSe_2$ vs. temperature at zero magnetic field and field $H_0=0.92T$ at different angles θ with respect to the (a,b) plane. (The units of $\Delta\chi'$ are $-1000\Delta f/f$, where f is the frequency of the RF tank circuit.) Curves are guides to the eye.

Fig.4 - dH/dT_{ON} vs. angle for $NbSe_2$ and NbS_2 crystals; curves are fits to equation (1) with mass anisotropies=8 ($NbSe_2$) and 64 (NbS_2).

in Figure 4. For an anisotropic superconductor, an effective mass model predicts [13]:

$$\frac{dH(\theta)}{dT_{ON}} \propto [\sin^2\theta + \frac{m_{a,b}}{m_c} \cos^2\theta]^{-1/2}. \quad (1)$$

The curves show fits to this equation, with $m_c/m_{a,b} = 8$ for NbSe_2 and 64 for NbS_2 . Because of the larger anisotropy of the sulfide, we expected the features below T_C to be more pronounced than for the selenide.

To our considerable surprise, no prominent features below T_U were observed in either magnetic measurements (Figure 5) or vibrating reed measurements (Figure 6) at any orientation. Unlike NbSe_2 , NbS_2 behaved like a simple, "conventional," superconductor; i.e. the frequency shift for both the vibrating reed and RF tank circuit varied monotonically with temperature and only a single peak was observed in vibrating reed dissipation.

Fig.5 - Effective ac susceptibility (the units of $\Delta\chi'$ are $-1000\Delta f/f$, where f is the frequency of the RF tank circuit) for a NbS_2 crystal at 90° and different fields; curves are guides to the eye.

Fig.6 - Resonant frequency shift and damping ($1/Q$) of a NbS_2 vibrating reed ($f \sim 8.3$ kHz) vs. temperature for $\theta=10^\circ$ and $\theta=45^\circ$.

It is therefore necessary to consider other differences between the selenide and sulfide crystals. Our crystals of NbSe_2 (RRR ~ 30) were less pure than the sulfide crystals (RRR ~ 115), but both materials are in the "clean" limit; the transport mean free paths (>300 Å for NbSe_2 [11]) are greater than Pippard coherence lengths (76 Å [11] and 140 Å [12], for NbSe_2 and NbS_2 , respectively). NbSe_2 has a well documented "peak" effect [14], in which the critical current (i.e. to depin the vortices) varies non-monotonically with field. While such has not been investigated in NbS_2 , the proposed explanations of this effect, involving an increase of pinning as the FLL shear modulus, C_{66} , decreases near T_C [14],

should also hold for the sulfide, and in any case, it is not clear how the critical state peak effect is related to the relatively smaller vortex motions we are probing. (The reed vibration amplitudes are typically $<100\text{\AA}$.)

Finally, in 2H-NbSe_2 , an incommensurate CDW with wavelength $9.0\text{\AA}$ coexists with the superconducting state [15], while there is no CDW in 2H-NbS_2 [12]. The CDW in NbSe_2 has an onset at 33K [15]; e.g. see Figure 7. We suggest that the softening in the FLL at $T_F(H)$ is caused by an interaction between the FLL and the CDW. This can occur if there is a change in the structure of the FLL at T_F , resulting in a change in Labusch parameter.

The fact that the frequency dips are strongest for $\hat{e}||H_0$ is consistent with a CDW/vortex interaction model, since the charge density modulations are in the (a,b) plane. That $T_F(H)$ and $T_{ON}(H)$ are approximately parallel (see Figure 2), rather than diverging at large H , implies that the interaction is not affected by the FLL spacing (which varies from $400\text{\AA}$ to $1700\text{\AA}$ for the fields we examined); the interaction must be with individual vortices. However, the hysteresis indicates that the softening is affected by the pinning configuration. In tunneling spectra of the sub-gap (i.e. vortex core) states of NbSe_2 , for $T \ll T_C$, Hess et al.[16] observed angle dependent structure that appeared to be related to the CDW. This structure was, however, spatially much larger than the Pippard coherence length, $76\text{\AA}$ [11]. As the coherence length grows near T_C , the effect of these states on the vortex pinning may change. It would therefore be

Fig.7 - Temperature dependence of ab-plane resistivities, normalized to room temperature values, of NbS_2 and NbSe_2 crystals. The bump near 30K for NbSe_2 is due to formation of the CDW.

interesting to search for changes in the FLL at T_F with imaging, but this may be impossible this close to T_C due to thermal broadening. A more feasible experiment may be to compare the spatial structure in the core tunneling spectra of $NbSe_2$ and NbS_2 .

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