Observation of Leggett’s collective mode in a multi-band MgB$_2$ superconductor

G. Blumberg$^{1,*}$, A. Mialitsin$^1$, B. S. Dennis$^1$, M. V. Klein$^2$, N. D. Zhigadlo$^3$, and J. Karpinski$^3$

$^1$Bell Laboratories, Alcatel-Lucent, Murray Hill, NJ 07974
$^2$Department of Physics and Materials Research Laboratory,
University of Illinois at Urbana-Champaign, Urbana, IL 61801
$^3$Solid State Physics Laboratory, ETH, CH-8093 Zürich, Switzerland

(Dated: March 30, 2007)

We report observation of Leggett’s collective mode in a multi-band MgB$_2$ superconductor with $T_c = 39$ K arising from the fluctuations in the relative phase between two superconducting condensates. The novel mode is observed by Raman spectroscopy at 9.4 meV in the fully symmetric scattering channel. The observed mode frequency is consistent with theoretical considerations based on the first principle computations.

PACS numbers: 74.70.Ad, 74.25.Ha, 74.25.Gz, 78.30.Er

The problem of collective modes in superconductors is almost as old as the microscopic theory of superconductivity. Bogolyubov $^{11}$ and Anderson $^{12}$ first discovered that density oscillations can couple to oscillations of the phase of the superconducting (SC) order parameter (OP) via the pairing interaction. In a neutral system these are the Goldstone sound-like oscillations which accompany the spontaneous gauge-symmetry breaking, however, for a charged system the frequency of these modes is pushed up to the plasma frequency by the Anderson-Higgs mechanism $^{13}$ and the Goldstone mode does not exist. The collective oscillations of the amplitude of the SC OP have a gap, which was first observed by Raman spectroscopy in NbSe$_2$ $^{14,15}$, and which plays a role equivalent to the Higgs particle in the electro-weak theory $^{16}$. Several other collective excitations have been proposed, including an unusual one that corresponds to fluctuations of the relative phase of coupled SC condensates first predicted by Leggett $^{17}$. The Leggett mode is a longitudinal excitation resulting from equal and opposite displacements of the two superfluids along the direction of the mode’s wavevector $q$. In the ideal case considered by Leggett, the mode is “massive” and its energy (mass) at $q = 0$ is below twice the smaller of the two gap energies. In this Letter we report the observation of Leggett’s collective mode in the multi-band superconductor MgB$_2$ with $T_c = 39$ K $^{18}$. The novel mode is observed in Raman response at 9.4 meV, consistent with the theoretical evaluations.

The multi-gap nature of superconductivity in MgB$_2$ was first theoretically predicted $^{19}$ and has been experimentally established by a number of spectroscopies. A double-gap structure in the quasi-particle energy spectra was determined from tunneling spectroscopy $^{20,21}$. The two gaps have been assigned by means of ARPES $^{22,23}$ to distinct Fermi surface (FS) sheets (Fig. 1) belonging to distinct quasi-2D $\sigma$-bonding states of the boron $p_{x,y}$ orbitals and 3D $\pi$-states of the boron $p_z$ orbitals: $\Delta_\sigma = 5.5 - 6.5$ and $\Delta_\pi = 1.5 - 2.2$ meV. Scanning tunneling microscopy (STM) has provided a reliable fit for the smaller gap, $\Delta_\pi = 2.2$ meV $^{24}$. This value manifests in the absorption threshold energy at 3.8 meV obtained from magneto-optical far-IR studies $^{25}$. The larger $2\Delta_\sigma$ gap has been demonstrated by Raman experiments as a SC coherence peak at about 13 meV $^{16}$.

Polarized Raman scattering measurements from the $ab$ surface of MgB$_2$ single crystals grown as described in $^{17}$ were performed in back scattering geometry using less than 2 mW of incident power focused to a $100 \times 200 \mu$m spot. The data in a magnetic field were acquired with a continuous flow cryostat inserted into the horizontal bore of a SC magnet. The sample temperatures quoted have been corrected for laser heating. We used the excitation lines of a Kr$^+$ laser and a triple-grating spectrometer for analysis of the scattered light. The data were corrected for the spectral response of the spectrometer and the CCD detector and for the optical properties of the material at different wavelengths as described in Ref. $^{18}$.

The factor group associated with MgB$_2$ is $D_{6h}$. We denote by $(e_{in},e_{out})$ a configuration in which the incoming/outgoing photons are polarized along the $e_{in}/e_{out}$ directions. The verti-

![FIG. 1: (color online) An illustration of the MgB$_2$ Fermi surface in the first Brillouin zone adapted from Ref. $^{23}$. A nearly cylindrical sheet of the FS around the $\Gamma - A$ line results from the $\sigma$-band. The $\pi$-band forms a FS of planar honeycomb tubular networks. For clarity only a single FS for each $\sigma$- and $\pi$-band pair is shown $^{29}$. In the SC state the $\sigma$-band Cooper pairs are bound stronger than the $\pi$-band pairs, at the binding energies $2\Delta_\sigma$ and $2\Delta_\pi$ correspondingly. Leggett’s collective mode originates from dynamic scattering of the $\sigma$-band pairs of electrons (illustrated in red) with momentum $(k, -k)$ into the $\pi$-band electron pairs (yellow) with momentum $(k', -k')$.](image-url)
BCS coherence peak singularity (shaded in dark and light green). The solid hairline above the shaded areas is the sum of both modes. To fit the observed shapes the theoretical gap cutoff, the SC coherence peak at

$$\Omega_{\text{coherence}}$$

while the peak in the visible range is associated with a transi-

tion from the interband contribution to the in-plane optical conductivity while both metal modes in the SC state (Fig. 2) experience renormalization below the SC transition [23]. For the $A_{1g}$ channel the spectra are dominated by two-phonon scattering.

### In Fig. 2 we show the Raman response from an MgB$_2$ single crystal for the $E_{2g}$ and $A_{1g}$ scattering channels for four excitation photon energies in the normal and SC states. Besides the phononic scattering at high Raman shifts all spectra show a moderately strong featureless electronic Raman continuum. The origin of this continuum is likely due to finite wave-vector effects [20, 24, 25]. For isotropic single band metals the Raman response in the fully symmetric channel is expected to be screened [20, 24, 26]. However, for MgB$_2$ the electronic scattering intensity in the $A_{1g}$ and $E_{2g}$ channels is almost equally strong.

The low frequency part of the electronic Raman continuum changes in the SC state (Fig. 2), reflecting renormalization of electronic excitations resulting in four new features in the spectra: (i) a threshold of Raman intensity at $2\Delta_0 = 4.6$ meV, (ii) a SC coherence peak at $2\Delta_1 = 13.5$ meV in the $E_{2g}$ channel, and two new modes in the $A_{1g}$ channel, (iii) at 9.4 meV, which is in-between the 2$\Delta_0$ and 2$\Delta_1$ energies, and (iv) a
much broader mode just below $2\Delta_1$. The observed energy scales of the fundamental gap $\Delta_0$ and the large gap $\Delta_1$ are consistent with $\Delta_\sigma$ and $\Delta_\pi$ as assigned by one-electron spectroscopies [12,13,14].

(i) At the fundamental gap value $2\Delta_0$ the spectra for both symmetry channels show a threshold without a coherence peak. This threshold is cleanest for the spectra with lower energy photon excitations $\Omega_{ex}$ for which the low-frequency contribution of multi-phonon scattering from acoustic branches is suppressed [23]. The absence of the coherence peak above the threshold is consistent with the expected behavior for a superconductor with SC coherence length larger than the optical penetration depth [24].

(ii) The $2\Delta_1$ coherence peak appears in the $E_{2g}$ channel as a sharp singularity with continuum renormalization extending to high energies, which agrees with expected behavior for clean superconductors [20,24,25]. The Raman coupling to this mode is provided by density-like fluctuations in the $\sigma$-band hence the peak intensity is enhanced by about an order of magnitude when the excitation photon energy $\Omega_{ex}$ is in resonance with the 2.6 eV $\sigma \rightarrow \pi$ inter-band transitions (Fig.3).

(iii) The novel peak at 9.4 meV is observed only in the $A_{1g}$ scattering channel. This mode is more pronounced for off-resonance excitation for which the electronic continuum above the fundamental threshold $2\Delta_0$ is weaker. We assign this feature to the collective mode proposed by Leggett [7]: If a system contains two coupled superfluid liquids a simultaneous cross-tunneling of a pair of electrons becomes possible (Fig 1). Leggett’s collective mode is caused by counter flow of the two superfluids leading to small fluctuations of the relative phase of the two condensates while the total electron density is locally conserved. In a crystalline superconductor, its symmetry is that of the fully symmetric irreducible representation of the group of the wave-vector $q$. If the energy of this mode is below the smaller pair-breaking gap energy, dissipation is suppressed and the excitation should be long-lived. In the case of MgB$_2$ the two coupled SC condensates reside at the $\sigma$- and $\pi$-bands. The oscillation between the con-

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|}
\hline
$\sigma_{\pi}$ & $\sigma_{\pi}$ & $\sigma_{\pi}$ & $\omega_L$ & $\omega_L$ & $\omega_L$ \\
\hline

\end{tabular}
\caption{Estimates of Leggett’s mode frequency $\omega_L$, the vertex correction $\omega_V$ and the Raman resonance frequency $\omega_{LR}$ based on values of intra- and inter-band pairing potentials $V_{ij}$ ($i,j=\pi,\sigma$) deduced from first principal calculations (two band model) [23]. The effective density of states $N_\sigma=2.04$ and $N_\pi=2.78$ Ry$^{-1}$ spin$^{-1}$ cell$^{-1}$ [23] and the experimental values for the SC gaps $\Delta_\sigma=6.75$ and $\Delta_\pi=2.3$ meV are used.}
\end{table}
Leggett's excitation $\gamma_\pi$ and $\gamma_\pi$ should not be equal, the coupling is further enhanced if $\gamma_\pi \gamma_\pi < 0$. The latter condition is satisfied for MgB$_2$ since the $\sigma$-bands are hole-like while the $\pi$-bands are predominantly electron-like. The integrated intensity of the Leggett's mode as a function of excitation energy does not follow the optical conductivity and is about five times weaker than the resonantly enhanced coherence peak in the $E_{2g}$ channel (Fig. 5).

The estimates of the two-band interaction matrices by first principle computations [8, 27, 28] which are collected in Table I show that for MgB$_2$ the condition (1) is satisfied. In Fig. 4 we show the calculated Raman response function (6) for the first set of parameters from Table I in the $q \rightarrow 0$ limit. Finite wave-vector contribution from the $\pi$-band will stretch the $\sigma$-band Raman continuum in agreement with the data. Model calculations suggest that interference with the $\sigma$-band coherence peak might produce a structure at about $2\Delta_\sigma$. We note that the estimates for bare Leggett's mode frequency $\omega_L$ are close to the $\sim 6.2$ meV value observed by tunneling spectroscopy [32] and the estimates for the peak in Raman response (6), $\omega_{LR}$, are consistent with the observed mode at 9.4 meV. Because the collective mode energy is between the two-particle excitation thresholds for $\pi$- and $\sigma$-band, $2\Delta_\pi < \omega_L < \omega_{LR} < 2\Delta_\sigma$, Leggett's excitation relaxes into the $\pi$-band continuum. Indeed, the measured $Q$-factor for this mode is about two: the mode energy relaxes into the $\pi$-band quasiparticle continuum within a couple of oscillations.

(iv) Finally we note that MgB$_2$ has four FSs, two nearly cylindrical sheets due to the $\sigma$-bands split and two tubular network structures originate from $\pi$-bands. Solution to the Leggett problem extended to 4-bands with $4 \times 4$ interaction matrix given by Liu et al. [9] leads to two Raman resonances: $\omega_{LR} = 8.4$ meV and second $\omega_{LR2}$ just 0.05 meV below the $2\Delta_t$ gap. We interpret the superconductivity induced intensity in the $A_{1g}$ channel just below the $2\Delta_t$ energy as evidence either for a second Leggett resonance or for interference between SC contributions from the $\pi$-band with large $qF_F$, and the $\sigma$-band with small $qF_F$. A sum of two modes peaking at 9.4 and 13.2 meV with very similar excitation profiles provides a good fit to the experimental data.

We conclude that despite being short lived, Leggett excitations in MgB$_2$ are observed in $A_{1g}$ Raman response.

The authors thank D. van der Marel, I. Mazin and W. E. Pickett for valuable discussions. AM was supported by the Lucent-Rutgers Fellowship program. NDZ was supported by the Swiss National Science Foundation through NCCR pool MaNEP.

![Figure 4: (color online) Im $\chi_{\Delta,\pi}(\omega)$ given by Eq. (6) using the interaction matrix by Liu et al. [9].](http://www.xcrysden.org/img/FS-viewing.png)